Methane production, recovery and emission from two Danish landfills

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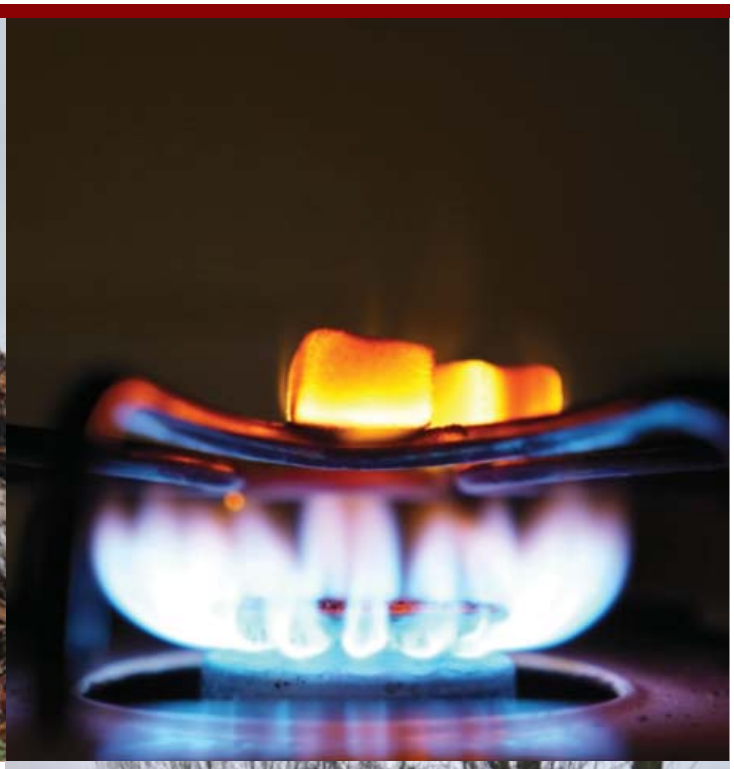

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PhD Thesis
April 2018

DTU Environment
Department of Environmental Engineering

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Preface

The work presented in this PhD thesis was carried out at the Department of Environmental Engineering of the Technical University of Denmark under the supervision of Professor Peter Kjeldsen and the co-supervision of Professor Charlotte Scheutz from January 2015 to April 2018 (including 15 weeks of parental leave). The PhD project was partially funded by the Odense Renovation A/S Company.

The thesis is organized in two parts: the first part puts into context the findings of the PhD in an introductive review; the second part consists of the papers listed below. These will be referred to in the text by their paper number written with the Roman numerals **I-III**.

I Aghdam, E.F., Scheutz, C., Kjeldsen, P., 2017. Assessment of methane production from shredder waste in landfills: The influence of temperature, moisture and metals. *Waste Management* 63, 226-237.

II Aghdam, E.F., Fredenslund, A.M., Chanton, J., Kjeldsen, P., Scheutz, C., 2018. Determination of gas recovery efficiency at two Danish landfills by performing downwind methane measurements and stable carbon isotopic analysis. *Waste Management* 73, 220-229.

III Aghdam, E.F., Scheutz, C., Kjeldsen, P., 2018. Impact of meteorological parameters on extracted landfill gas composition and flow. *Waste Management*, in press. doi:10.1016/j.wasman.2018.01.045.

In this online version of the thesis, papers I-III are not included, but can be obtained from electronic article databases, e.g., via www.orbit.dtu.dk, or on request from DTU Environment, Technical University of Denmark, Bygningstorvet, Building 115, 2800 Kgs. Lyngby, Denmark, info@env.dtu.dk.

In addition, the following publications, not included in this thesis, were also concluded during this PhD study:

Aghdam, E.F, Scheutz, C., Kjeldsen, P., 2016. Impact of meteorological parameters on extracted methane concentration at landfills. *Global Waste Management Symposium 2016*, California, United States.

Aghdam, E.F, Fredenslund, A. M., Kjeldsen, P. Scheutz, C., 2016. Quantification of methane emissions from two Danish landfills. *Sustain-ATV Conference 2016*, Kgs. Lyngby, Denmark.

Aghdam, E.F, Scheutz, C., Kjeldsen, P., 2017. The role of metals in methane production from shredder waste in landfills. *The 5th International Conference on Sustainable Solid Waste Management,* Athens, Greece.

Aghdam, E.F, Fredenslund, A. M., Kjeldsen, P., Scheutz, C., 2017. Gas collection efficiency at two Danish landfills. *Proceedings Sardinia 2017, Sixteenth International Waste Management and Landfill Symposium,* S. Margherita di Pula, Cagliari, Italy.

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I would also thank my parents and my little brother and sister for their support and love, and for encouraging me to move abroad and follow my dreams. I am very grateful for having such an understanding and kind family. I would especially like to thank my wife, Jenni, for quitting her job and moving to Denmark with me: you are amazing. I really could not do it without your love and support. Last but definitely not least, I thank our new-born daughter, Sofia. Your birth was certainly the best part of my PhD and writing the thesis was much easier by having you on the journey.

Summary

Landfill gas (LFG), mainly consisting of methane (CH_4) and carbon dioxide (CO_2), is produced by the anaerobic digestion of biodegradable waste deposited in landfills. CH_4 is a greenhouse gas with global warming potential 28 times that of CO_2 over a period of 100 years. The produced CH_4 in landfills can be recovered and utilized for the production of electricity and/or heat. Higher recovery of CH_4 could result in lower CH_4 emissions into the atmosphere, and thus lower the contribution of landfills to climate change. Moreover, higher CH_4 recovery can result in higher production of heat and electricity, leading to higher revenue for landfill owners. Therefore, it is important to understand the factors that can affect CH_4 recovery from landfills.

The amount of CH_4 recovered from a landfill is indeed a function of the amount of CH_4 produced in a landfill. The amount of produced CH_4 in a landfill is governed by the waste composition and can be affected by many factors, including temperature, moisture, and chemical or microbial reactions, which occur simultaneously inside a landfill, such as corrosion. Moreover, the amount of CH_4 recovered from a landfill depends on the efficiency of the gas recovery system, which is affected by its design and management, as well as the presence and type of top cover at the landfill. Furthermore, CH_4 recovery from a landfill can be affected by changes in meteorological parameters. For instance, changes in barometric pressure affect the pressure gradient, which is the driving force for advective gas transport, between inside the landfill and the atmosphere, and thus potentially can impact CH_4 recovery.

The overall goal of this PhD project was to address specific challenges regarding CH_4 production and recovery at landfills. The PhD project focused on three topics: 1) an in-depth investigation of CH_4 production from shredder waste (SW) at landfills, 2) the determination of gas recovery efficiency at two adjacent Danish landfills by field measurement, and 3) the influence of meteorological parameters on gas recovery from landfills. This PhD project focused on two adjacent Danish landfills, Stige Ø and Odense Nord, which are connected to the same gas recovery system.

In order to assess the CH_4 production from SW at landfills, SW was sampled from the Odense Nord landfill, size-reduced and characterized in terms of total solids (TS), volatile solids (VS), total carbon (TC), total organic carbon

(TOC) and biogenic carbon (BioC). SW samples were incubated to measure their first-order decay kinetic constant (k-value), under different operating conditions (temperature and moisture), and their biochemical methane potential (BMP). In addition, four main metals present in SW (Fe, Al, Zn and Cu) were examined for their ability to produce available H_2 for methanogens.

The characterization results showed that a high fraction of the organic carbon (47-61%) in SW is fossil carbon. Moreover, high TS content (82-91%) in the waste samples showed that the samples were fairly dry. The measured BMPs were 1.5-6.2 kg CH_4/ton waste, while the measured k-values were 0.033-0.075/yr at room temperature, 0.220-0.429/yr at 37 ºC and 0.235-0.488/yr at 55 ºC. The fine fraction of SW obtained after sieving showed a lower BMP and k-value in comparison to the unsieved SW, meaning that landfilling of the fine fraction of SW could result in lower CH_4 production in comparison to unsieved SW.

Carrying out the incubation experiments under different operating conditions indicated the high dependency of the CH_4 production rate on temperature and moisture. H_2 was produced by biocorrosion of Fe, Al and Zn and utilized by methanogens to convert CO_2 into CH_4. The addition of Al and Zn to the incubated SW resulted in higher CH_4 production. Relatively high CH_4 production from SW at landfills and the unusual gas composition (high CH_4 and low CO_2 content) are most likely due to methanogens converting the existing CO_2 in the produced LFG into CH_4, using the H_2 produced by biocorrosion of Al and Zn.

In order to determine the gas recovery efficiency at the landfills, a set of field activities was performed: whole-site CH_4 emissions were measured by the tracer gas dispersion method (six measurement campaigns), while CH_4 oxidation in the top layer of the landfills was measured by stable carbon isotopic analysis (two measurement campaigns). The CH_4 recovery rate, which was provided by landfill operators, was the sum of the CH_4 recovered from both landfills. In addition, the total CH_4 production rate was estimated using the Afvalzorg model and compared with the results of field measurements.

Total CH_4 emissions from the two landfills combined were 29-50 kg/h, while the measured CH_4 oxidation efficiency was 6-37%. The CH_4 recovery rate from both landfills combined was 85-115 kg/h. The calculated gas recovery efficiency was 59-76%, which indicated a high potential for improvement in the gas recovery system at landfills. The average total CH_4 production rate

determined by field measurements (sum of methane recovered, emitted and oxidized) was 147 kg/h, which was close to the estimated total CH_4 production rate of 154 kg/h by the Afvalzorg model. The calculated gas recovery efficiency, along with the observed major CH_4 emission areas during the surface screenings, can be used for developing a plan for improvement of the gas recovery system.

In order to investigate the influence of meteorological parameters on LFG recovery, correlation coefficients and p-values were calculated between the gas recovery data and meteorological parameters. Barometric pressure, wind speed, ambient temperature and solar radiation were the chosen meteorological parameters. Four periods (from a few days to approximately one month) in 2015 and 2016 were studied. These four periods were chosen because the gas recovery system was not manually adjusted in these periods by the landfill operators.

Relatively high correlation coefficients were observed between LFG data (LFG CH_4 concentration, LFG flow and CH_4 flow) and barometric pressure ($|r| = 0.37\text{-}0.73$), while higher correlation coefficients were observed between LFG data and changes in barometric pressure ($|r| = 0.80\text{-}0.93$). Moreover, a strong correlation was observed between wind speed and LFG data in winter ($|r| = 0.75\text{-}0.77$), but not in summer ($|r| = 0.05\text{-}0.30$).

The correlations of LFG data with barometric pressure, changes in barometric pressure and wind speed were statistically significant ($p < 0.01$) and observed visually in scatterplots. The slope of the linear regression between changes in barometric pressure and LFG data can be used to predict changes in recovered LFG as a function of changes in pressure. As LFG recovery data only correlated weakly with ambient temperature ($|r| = 0.12\text{-}0.49$) and solar radiation ($|r| = 0.03\text{-}0.21$), these two parameters were not found to affect LFG recovery.

Dansk sammenfatning

Deponigas, som primært består af metan (CH_4) og kuldioxid (CO_2), dannes ved anaerob nedbrydning af bionedbrydeligt affald i deponier. CH_4 er en drivhusgas med et drivhusgaspotentiale 28 gange større end CO_2 målt over en periode på 100 år. CH_4 dannet i deponier kan indvindes og nyttiggøres til produktion af elektricitet og/eller varme. En større indvinding af CH_4 kan give en lavere CH_4-udledning til atmosfæren og dermed minimere deponiernes bidrag til den globale opvarmning. Desuden kan en bedre CH_4-indvinding give en højere produktion af varme og elektricitet, som igen vil bidrage med højere indtægter til deponiejerne. Derfor er det vigtigt at forstå de faktorer, der kan påvirke nyttiggørelsen af CH_4 fra deponier.

Mængden af indvundet CH_4 fra et deponi er en funktion af den genererede CH_4-mængde i deponiet. Mængden af genereret CH_4 i et deponi afhænger af affaldssammensætningen og kan påvirkes af mange faktorer, blandt andet temperatur, fugt og kemiske eller mikrobielle reaktioner, som sker samtidigt inde i deponiet. Af kemiske reaktioner kan nævnes korrosion af metaller. Endvidere er mængden af indvundet CH_4 fra deponiet afhængig af effektiviteten af gasindvindingssystemet, som igen er afhængig af, hvordan systemet er designet, og hvordan det drives samt hvilken type slutafdækning, der er på deponiet. Der ud over kan CH_4-indvindingen fra deponiet blive påvirket af meteorologiske forhold. For eksempel vil ændringer i barometertrykket påvirke trykgradienten, som er den drivende kraft for advektiv gastransport mellem det indre af deponiet og atmosfæren. Dette kan potentielt påvirke CH_4-indvindingen.

Det overordnede mål for dette PhD projekt har været at adressere specifikke udfordringer i forhold til CH_4-dannelsen og -indvindingen fra deponier. PhD projektet har fokuseret på tre emner: 1) en dybdegående undersøgelse af CH_4-dannelsen i shredderaffald på deponier, 2) bestemmelse af gasindvindingseffektiviteten på to danske deponier ved brug af feltmålinger og 3) effekten af meteorologiske forhold på gasindvindingen fra deponier. I dette PhD-projekt er der fokuseret på de to deponier, Stige Ø og Odense Nord, som begge er koblet på det samme gasindvindingssystem.

For at kunne vurdere CH_4-dannelsen i shredderaffald, blev der indsamlet prøver af shredderaffald fra deponiet Odense Nord. Prøverne blev neddelt og karakteriseret i forhold til indholdet af tørstof (TS), flygtige stoffer (VS), totalt kulstof (TC), totalt organisk kulstof (TOC) og biogent kulstof (BioC).

Prøverne med shredderaffald blev inkuberet for at bestemme første-ordens-nedbrydningskonstanten (k-værdien) under forskellige forhold (temperatur og fugt) og det biokemiske CH_4 potentiale (BMP). Der ud over blev indholdet af de fire væsentligste metaller i shredderaffaldet (Fe, Al, Zn and Cu) bestemt samt disse metallers evne til at danne H_2, som kan være basis for metandannende reaktioner udført af methanogene bakterier.

Resultaterne af karakteriseringen viste, at en stor del af det organiske kulstof (47-61%) i shredderaffald er fossilt kulstof. Endvidere viste TS indholdet (82-91%), at affaldsprøverne var relativt tørre. Det målte BMP var 1,5-6,2 kg CH_4/ton affald, mens den målte k-værdi var 0,033-0,075/år ved rumtemperatur, 0,220-0,429/år ved 37 °C og 0,235-0,488/år ved 55 °C. Den fine fraktion af shredderaffaldet, som fås ved at sigte affaldet, viste lavere BMP og k-værdi, hvilket betyder, at deponering af den fine fraktion af shredderaffald kan resultere i en lavere CH_4-produktion i forhold til ikke-sigtet shredderaffald.

Gennemførelsen af inkubationsforsøget under forskellige forhold indikerede, at CH_4-produktionsraten var påvirket af temperaturen og vandindholdet. H_2 blev dannet ved biokorrosion af Fe, Al og Zn og blev udnyttet af methanogene bakterier ved omdannelse af CO_2 til CH_4. Tilsætning af Al og Zn til det inkuberede shredderaffald resulterede i en højere CH_4-produktion. Den relativt høje CH_4-produktion fra shredderaffald i deponier og den usædvanlige gassammensætning (højt CH_4- og lavt CO_2-indhold) skyldes højst sandsynligt, at metanogene bakterier omdanner CO_2 i den genererede deponigas til CH_4 ved hjælp af H_2 dannet ved biokorrosion af Al og Zn.

For at kunne bestemme gasindvindingseffektiviteten fra deponierne blev der gennemført en række feltundersøgelser. Den samlede CH_4-udledning blev målt ved brug af sporgasmetoden (seks målekampagner), mens CH_4-oxidationen i slutafdækningen på deponierne blev målt ved analyse af stabile kulstof isotoper (to målekampagner). CH_4-indvindingsrater blev oplyst af de driftsansvarlige på deponierne som en sum af CH_4-indvindingen fra begge deponier. Desuden blev den totale CH_4-produktionsrate estimeret ved brug af en gasproduktionsmodel (Afvalzorg-modellen) og sammenlignet med resultaterne af feltmålingerne.

Den samlede CH_4-udledning fra de to deponier blev målt til 29-50 kg/h, mens den målte CH_4-oxidationseffektivitet var 6-37 %. CH_4-indvindingsraten fra begge deponier tilsammen var 85-115 kg/h. Den beregnede gasindvindingseffektivitet var 59-76 %, hvilket indikerede et stort

forbedringspotentiale for gasindvindingssystemet for deponierne. Den gennemsnitlige totale CH_4-produktionsrate bestemt ved feltmålinger (summen af den indvundne, udledte og oxiderede metan) var 147 kg/h, hvilket var tæt på den estimerede totale produktionsrate på 154 kg/h beregnet i Afvalzorg-modellen. Den beregnede gasindvindingseffektivitet sammenholdt med de væsentligste observerede CH_4-udledningsområder fundet ved overfladescreening kan bruges til udarbejdelse af en plan for forbedring af gasindvindingssystemet.

For at undersøge betydningen af meteorologiske forhold for deponigasindvindingen, blev korrelationskoefficienter og p-værdier beregnet for forholdet mellem gasindvindingsdata og meteorologiske forhold. De valgte meteorologiske forhold var barometertryk, vindhastighed, lufttemperatur og solindstrålingen. Fire perioder (fra nogle få dage til ca. en måneds varighed) i 2015 og 2016 blev valgt til sammenligningen. Disse fire perioder var valgt, fordi gasindvindingssystemet ikke blev manuelt justeret af driftspersonalet i disse perioder.

Relativt høje korrelationskoefficienter mellem deponigasdata (koncentrationen af CH_4 i deponigassen, deponigasflowet og CH_4-flowet) og barometertrykket blev fundet ($|r| =$ 0,37-0,73), mens endnu højere korrelationskoefficienter blev fundet mellem deponigasdata og ændringer i barometertrykket ($|r| =$ 0,80-0,93). I øvrigt blev der fundet en stærk korrelation mellem vindhastighed og deponigasdata om vinteren ($|r| =$ 0,75-0,77) men ikke om sommeren ($|r| =$ 0,05-0,30).

Korrelationen mellem deponigasdata og barometertryk, ændringer i barometertryk og vindhastighed var statistisk signifikant ($p < 0.01$) og kunne observeres visuelt i korrelationsplots. Hældningen på den lineære regression mellem ændringer i barometertryk og deponigasdata kan bruges til at forudsige ændringer i indvundet deponigas som en funktion af ændringer i trykket. Da deponigasindvindingsdata kun havde en svag korrelation til lufttemperatur ($|r| =$ 0,12-0,49) og solindstråling ($|r| =$ 0,03-0,21) vurderes det, at disse to forhold ikke påvirkede indvindingen af deponigas.

Table of contents

Preface ..... **i**

Acknowledgements ..... **iii**

Summary ..... **iv**

Dansk sammenfatning ..... **vii**

Table of contents ..... **x**

Abbreviations ..... **xii**

1 Introduction **1**

1.1 Background 1

1.2 Research objectives 3

2 Material and methods **5**

2.1 Landfills site description 5

2.2 Methane production from shredder waste at landfills 6

2.2.1 Waste sampling and characterization 6

2.2.2 Biochemical methane potential 7

2.2.3 Biocorrosion experiment 7

2.2.4 Gas production rate experiment 8

2.3 Determination of gas recovery efficiency at the landfills 10

2.3.1 Methane recovery measurements 10

2.3.2 Methane emission measurements 11

2.3.3 Methane oxidation measurements 12

2.3.4 Methane lateral migration 14

2.3.5 Changes in methane storage 14

2.3.6 Methane production modelling 14

2.4 Impact of meteorological parameters on methane extraction from the landfills 15

2.4.1 Data collection 15

2.4.2 Statistical analysis 15

3 Results and discussion **17**

3.1 Methane production from shredder waste at landfills 17

3.1.1 Waste characterization 17

3.1.2 Biochemical methane potential 17

3.1.3 Biocorrosion experiment 18

3.1.4 Gas production rate experiment 19

3.2 Determination of gas recovery efficiency at the landfills 20

3.2.1 Methane emission measurements 20

3.2.2 Methane oxidation measurements 22

3.2.3 Total methane production rate and gas recovery efficiency 23

3.2.4 Modelling and comparison with field measurements 25

3.3 Impact of meteorological parameters on methane extraction from the landfills 26
3.3.1 Barometric pressure and changes in barometric pressure 26
3.3.2 Ambient temperature 30
3.3.3 Wind speed 32
3.3.4 Solar radiation 35
4 Conclusions and perspectives 37
5 Future research 38
6 References 39
7 Papers 44

Abbreviations

Al	Aluminium
BioC	Biogenic carbon
BMP	Biochemical methane potential
CH_4	Methane
CO_2	Carbon dioxide
Cu	Copper
Fe	Iron
FOD	First-order decay
FS	Fresh sieved
COM	Composite sample
WWTP	Wastewater treatment plant
FUS	Fresh unsieved
GRE	Gas recovery efficiency
H_2	Hydrogen
LFG	Landfill gas
MPR	Measuring, pump and regulation
NTP	Normal temperature and pressure
SW	Shredder waste
SW2009	Shredder waste from 2009
SW2012	Shredder waste from 2012
TC	Total carbon
TOC	Total organic carbon
TS	Total solids
VS	Volatile solids
Zn	Zinc

1 Introduction

1.1 Background

Anaerobic digestion of waste containing biodegradable organic matter in landfills results in landfill gas (LFG) production, which consists of about 55-60% v/v methane (CH_4) and 40-45 % v/v carbon dioxide (CO_2). Emissions from landfills contribute to global warming as CH_4 is a greenhouse gas with global warming potential 28 times that of CO_2 (IPCC, 2013). Apart from contributing to global warming, the produced LFG can result in explosion, fire, odour and vegetation damage, if not managed properly (Christensen, 2011).

Landfilling of organic waste has been banned since 1997 in Denmark, due to the negative environmental impacts of landfills. Since then, waste types with low organic carbon content have been deposited in Danish landfills, for instance, shredder waste (SW). SW is the residual fraction after mechanical treatment and metal recovery from discarded vehicles and separately collected metal-containing waste, such as white goods, bicycles and strollers.

SW mostly consists of plastic, rubber, metals and wood (Fiore et al., 2012). High CH_4 production from SW monofills has been reported previously (Mønster et al., 2015; Scheutz et al., 2011b), even though it contains low biodegradable fractions. Moreover, previous studies have shown that the produced LFG in SW monofills has higher CH_4 and lower CO_2 contents in comparison to conventional LFG (Olsen and Willumsen, 2013; Scheutz et al., 2011b). Furthermore, high temperatures (59 and 40 °C at 10- and 20-m depths) have been observed previously inside SW monofills (Olsen and Willumsen, 2013). The reasons for high CH_4 production, unusual gas composition and high temperatures at SW monofills are unknown.

Iron (Fe), aluminium (Al), zinc (Zn) and copper (Cu) are the most abundant metals found in SW (Ahmed et al., 2014; Fiore et al., 2012; Granata et al., 2011). H_2 can be produced by the corrosion of metals under anaerobic conditions (Belay and Daniels, 1990; Hu et al., 2015; Lorowitz et al., 1992). In anaerobic digestion process, H_2 can be used by methanogens to convert CO_2 into CH_4 (Wise et al., 1978). The high CH_4 production and unusual gas composition at SW monofills could be due to H_2 production by corrosion of metals present in SW and the utilization of the produced H_2 by methanogens to convert the existing CO_2 in the produced biogas into CH_4, resulting in higher CH_4 and lower CO_2 content of the produced gas.

Despite the ban on the landfilling of organic waste since 1997, many Danish landfills (e.g., old landfills containing organic waste and SW monofills) still continue to produce CH_4. At some landfills, gas extraction facilities are installed, where it is affordable. The recovered CH_4 can be used as a renewable energy source for the production of heat and electricity. Higher CH_4 recovery rates result in lower CH_4 emissions into the atmosphere and can generate revenue by the higher production of heat and electricity. Thus, improving the gas recovery system is beneficial, both environmentally and economically.

In order to assess the potential for improvement of the gas recovery system, the gas recovery efficiency (GRE) firstly needs to be determined. A range of GRE determinations by field measurements is reported in the literature (Börjesson et al., 2009; Lohila et al., 2007; Mosher et al., 1999; Spokas et al., 2006). The GRE can be determined by dividing the CH_4 recovery rate by the total CH_4 production rate, which can be calculated using the following equation (Bogner and Spokas, 1993):

$$CH_4\ produced = CH_4\ emitted + CH_4\ oxidized + CH_4\ recovered + CH_4\ migrated + \Delta CH_4\ storage \quad \text{(Eq. 1)}$$

A gas recovery system can collect only a portion of the total produced CH_4. A portion of the produced CH_4 is emitted into atmosphere. There are different methods available for the quantification of CH_4 emissions from landfills, such as using flux chambers (Barlaz et al., 2004; Christophersen et al., 2001), micrometeorological measurements (Lohila et al., 2007; McBain et al., 2005) and the tracer gas dispersion method (Börjesson et al., 2009; Scheutz et al., 2011c; Mønster et al., 2015; 2014).

A portion of the produced CH_4 is oxidized into CO_2 by methanotrophs when passing through the top cover of a landfill. Stable carbon isotopic analysis, based on the preference of methanotrophs to oxidize ^{12}C faster than ^{13}C, is a method that has been used for the determination of CH_4 oxidation at landfills (Börjesson et al., 2009; Chanton et al., 1999).

A portion of the produced CH_4 can migrate off-site, which can be minimized by the bottom lining of landfills using geomembrane or geosynthetic clay (Spokas et al., 2006). A portion of the produced CH_4 is stored in the body of the landfill. The amount of the stored CH_4 is affected by several factors, including changes in barometric pressure and the moisture content of the cover (Scheutz et al., 2009).

In order to improve CH_4 recovery from landfills, it is important to understand the factors that can affect the recovery rate and LFG composition. Meteorological parameters can impact CH_4 production and emission at landfills (Scheutz et al., 2009), and thus could potentially impact the CH_4 recovery. There are relatively few studies reported in the literature that have focused on the impact of meteorological parameters on the recovered LFG composition and flow (Paper **III,** Table 1).

Czepiel et al. (2003), for instance, found that measured CH_4 emissions correlated with the absolute value of barometric pressure, while other studies (Fredenslund et al., 2010; Gebert and Groengroeft, 2006; Nastev et al., 2001; Poulsen et al., 2003; Xu et al., 2014) found that changes in barometric pressure affect LFG emissions. Czepiel et al. (2003) reported no correlation between ambient temperature and CH_4 emissions at a US landfill, while Christophersen et al. (2001) reported the significant impact of ambient temperature on LFG emissions at a Danish landfill.

There are also few studies available in the literature that have focused on the impact of wind speed (Poulsen, 2005; Xin et al., 2016), wind direction (Xu et al., 2014) and solar radiation (Xin et al., 2016) on LFG emissions. To the best of our knowledge, the majority of studies have focused on the impact of meteorological parameters on LFG emissions, but not on LFG recovery. Thus, there is a need to further investigate the impact of these parameters on the recovered LFG.

1.2 Research objectives

The overall objective of this study was to address some of the challenges at new and old landfills regarding CH_4 production and recovery. The specific objectives of the study were as follows:

- To investigate the ability of metals in SW to produce H_2, which could be used by methanogens to convert CO_2 into CH_4. In addition, the impact of sieving, temperature, moisture and inoculum addition on CH_4 production from SW was assessed. Moreover, the biochemical methane potential (BMP) and k-value of SW were determined. These objectives were met by performing different batch incubation experiments under different operating conditions.
- To determine the GRE of the two adjacent Danish landfills by measuring the CH_4 emission rate (using the tracer gas dispersion method), CH_4 oxidation rate (by stable carbon isotopic analysis) and CH_4 recovery rate

(recorded at the power plant of the landfills). In addition, the total CH_4 production at the landfills was modelled and compared with the results of the field measurements.

- To investigate the impact of selected meteorological parameters (barometric pressure, wind speed, ambient temperature and solar radiation) on the recovered LFG flow and composition by performing statistical correlation tests and a visual check of correlations in scatterplots.

2 Material and methods

2.1 Landfills site description

This PhD study focused on two adjacent Danish landfills: Stige Ø and Odense Nord, located in Odense, Denmark. Figure 1 shows a map of these landfills. The Stige Ø was established in 1967 and in operation until 2005. This landfill received different types of waste generated in Odense until 1994, after which it received only soil until its closure. No bottom liner is in place at the Stige Ø landfill. However, a leachate collection system has been installed at the landfill. The leachate is pumped into the wastewater treatment plant (WWTP) located at the Odense Nord landfill.

The Stige Ø landfill covers an area of 56 ha. The majority of the Stige Ø surface is covered with 1 m of soil. In some areas, the cover is slightly thicker due to construction work by the Odense municipality to convert the landfill into a recreational area with different facilities for outdoor activities after its closure. In total, around 7 million tons of waste and soil have been deposited in this landfill.

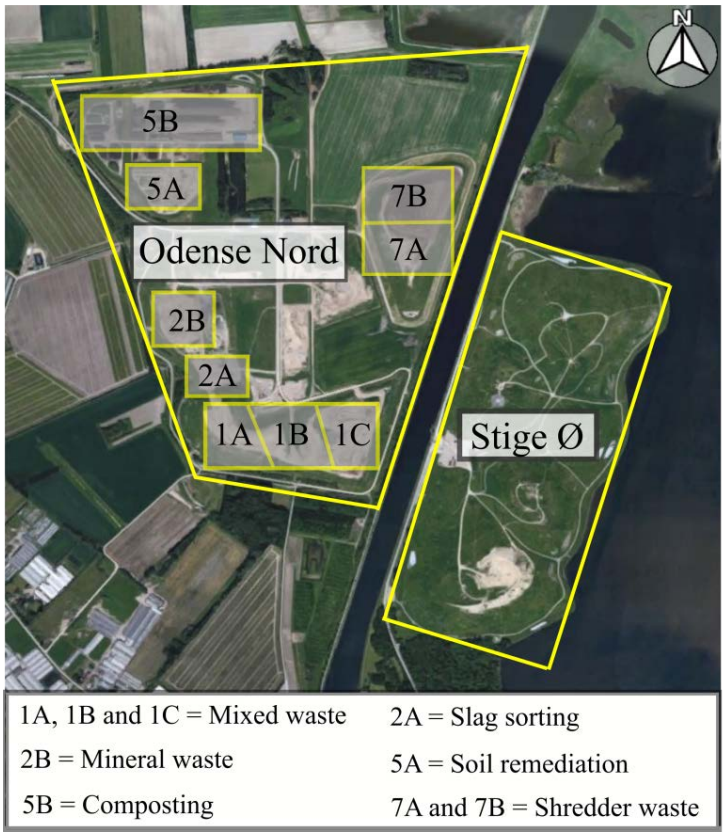

Figure 1. Map of Stige Ø (right) and Odense Nord (left) landfills (Imagery ©2016 Google, Aerodata International Surveys).

The Odense Nord landfill was established in 1994 and is still in operation. The landfill receives different types of waste, including SW, mineral waste and mixed waste. Apart from landfilling, soil remediation and composting activities (cells 5A and 5B, respectively) take place at the north-west part of the site. In the composting facility of the site, garden waste, sewage sludge and straw are co-composted.

In the eastern part of the Odense Nord landfill, SW is deposited (Cell 7). The SW cell covers an area of 6.5 ha and comprises two sections (7A and 7B). Both sections have reached their maximum capacity, but have not been finally covered due to potential landfill mining of the waste in the near future. In the southern part of the landfill, mixed waste is deposited over an area of 12.6 ha (Cell 1) and comprises three sections (1A, 1B and 1C). Two sections (1A and 1B) are finalized and covered with 1-10 m of soil, and the third (1C) is still in operation. The bottom lining system in the SW and mixed waste cell is composed of a 1-mm HDPE membrane on top of a 30-cm clay layer. A leachate collection system has been installed at the Odense Nord landfill. The leachate is pumped into the WWTP located at the site.

An active gas recovery system, consisting of 216 gas wells, is in place in the Stige Ø landfill and the SW (Cell 7) and mixed waste cells (Cell 1) of the Odense Nord landfill. At the Stige Ø landfill, there are 160 vertical gas wells. In the mixed waste cell of the Odense Nord landfill, there are 19 vertical and 10 horizontal gas wells, while there are 13 vertical gas wells installed in one section of the SW cell (7A). In addition, 14 vertical gas wells were installed in the second section of the SW cell (7B) on May 2016. The gas wells are connected to six measuring, pump and regulation modules (MPR modules), which are connected to a local power plant where a gas engine and a boiler produce electricity and heat.

2.2 Methane production from shredder waste at landfills

2.2.1 Waste sampling and characterization

SW samples were taken from the Odense Nord landfill, according to the year when they were landfilled: 2009 (SW2009), 2012 (SW2012) and fresh samples in 2015 (FUS). A part of the FUS sample was sieved with a drum sieve with a mesh size of 10 mm at the site, which is referred to as fresh sieved (FS). The sampling procedure is described in detail in Paper **I**. The SW2009, SW2012 and FUS were mixed together, based on equal wet weight,

in order to produce a composite sample (COM) that was representative of the SW disposed of in the landfill. Then the total solid (TS) content of the samples was measured according to Standard Methods (APHA, 2005).

Metals, wires and stones were separated manually from 5 kg of each sample, which was then size reduced with a cutter mill using a 1-mm sieve. The size-reduced samples were characterized in terms of volatile solids (VS), total carbon (TC), total organic carbon (TOC) and biogenic carbon (BioC) content. Analysis of TS and VS were conducted according to Standard Methods (APHA, 2005). TC and TOC were analysed according to EN 13137 (2001), while BioC content of the samples was determined according to EN 15440 (2011).

2.2.2 Biochemical methane potential

A BMP test was conducted on the size-reduced SW2009, SW2012, FUS, FS and COM samples at 37 °C in 1-L glass bottles. The experiment was performed in triplicate and lasted for 37 days, in which the organic loading was set to 5 g VS/L, while the inoculum to substrate ratio was 1 g VS/g VS. Iron particles of 5 mm × 2.5 mm were added to the SW2009 in order to investigate the impact of the metals, which were removed from the samples before size reduction. The reactors were flushed with N_2 for 15 min in order to create anaerobic condition and then sealed with a rubber septum.

Inoculum was collected from a biogas reactor located at VA Syd Sjölunda WWTP (Malmö, SE). Bottles containing only inoculum and water were used as blanks. The CH_4 produced from the blank experiment was subtracted from the CH_4 production of substrates to calculate the CH_4 production of the substrates alone. Control experiments, with Avicel as the standard substrate, were conducted in order to validate the experiment. Concentration of CH_4 in the headspace of the reactor was determined with a trace gas chromatograph (TRACE 1310 GC, Q PLOT, 0.32 mm, 8 m).

The theoretical CH_4 potential of the substrates was calculated by assuming that all organic carbon of the substrates could be degraded to an equal volume fraction of CH_4 and CO_2 (Paper **I**, Eq. 6). The CH_4 recovery was determined by dividing the CH_4 production in the BMP experiment by the calculated theoretical CH_4 production.

2.2.3 Biocorrosion experiment

Biocorrosion experiment was conducted at 37 °C in 1-L glass bottles for 20 days under anaerobic conditions. This experiment had two steps. In the first

step, Fe, Al, Zn and Cu were checked for their ability to produce H_2. In this step, these elemental metals (2.5 g of each) were placed in contact with water (250 g) in incubation bottles, resulting in a concentration of 10 g/L of liquid for each metal. In the second step, we investigated whether CH_4 production from inoculum could be enhanced by the addition of Fe, Al, Zn and Cu. In this step, 2.5 g of each metal was placed in contact with 250 g of inoculum.

Bottles containing only inoculum (blanks) were used to calculate the CH_4 production from inoculum alone. Inoculum collected from VA Syd Sjölunda WWTP was used in this experiment. Concentrations of CH_4 and H_2 in the headspace of the reactors were determined using a trace gas chromatograph (TRACE 1310 GC, Molsieve, 0.53 mm, 30 m). The elemental metals used in this experiment were in powder form and obtained from Sigma-Aldrich.

2.2.4 Gas production rate experiment

This experiment was conducted in 5-L glass bottles, connected to 2-L aluminium gas bags with FS, FUS and COM as substrates for 230 days under anaerobic conditions. In addition, elemental metals (Fe, Al, Zn and Cu) with dimension of 5 mm × 5 mm and concentrations of 25, 10, 10 and 10 g/L of liquid, respectively, were added to the reactors containing the COM sample in order to investigate whether the addition of metals could enhance the CH_4 production of these reactors, in comparison to the reactor containing only the COM sample. We also conducted abiotic experiments with sterilized samples (autoclaving three times for 1 h at 121 °C), in order to investigate whether there was any non-microbial production of CH_4 from SW.

Table 1 shows an overview of the gas production rate experiment. This experiment had two phases. During the first 100 days (Phase I), the experiments were conducted at room temperature in triplicate. The moisture content was adjusted to 35% w/w in all reactors, and inoculum (5% of substrate wet weight) was added to the biological rectors.

However, because of very low CH_4 production, the reactors were opened on day 100, more inoculum (30% of substrate wet weight) was added and the moisture content was adjusted to 75% w/w (Phase II, Days 100-230). Moreover, in this phase, the reactors were placed at two different temperatures, 37 and 55 °C (one replicate at each temperature) and one replicate remained at room temperature. Figure 2 shows the set-up of the experiment in Phase I when all reactors were incubated at room temperature.

Table 1. Overview of the conditions and phases of the gas production rate experiment.

Name of the reactor	Biotic/ abiotic	Inoculum addition (% of the samples wet weight)		Temperature (ºC)		Moisture content (% w/w)	
		Phase I	Phase II	Phase I	Phase II	Phase I	Phase II
FS1, FUS1, COM1, COM+Fe1, COM+Al1, COM+Zn1, COM+Cu1	Biotic	5	30	20-25	20-25	35	75
Ste_COM1[(a)], Ste_COM+Fe1, Ste_COM+Al1, Ste_COM+Zn1, Ste_COM+Cu1	Abiotic	0	0				
FS2, FUS2, COM2, COM+Fe2, COM+Al2, COM+Zn2, COM+Cu2	Biotic	5	30	20-25	37	35	75
Ste_COM2, Ste_COM+Fe2, Ste_COM+Al2, Ste_COM+Zn2, Ste_COM+Cu2	Abiotic	0	0				
FS3, FUS3, COM3, COM+Fe3, COM+Al3, COM+Zn3, COM+Cu3	Biotic	5	30	20-25	55	35	75
Ste_COM3, Ste_COM+Fe3, Ste_COM+Al3, Ste_COM+Zn3, Ste-COM+Cu3	Abiotic	0	0				

[(a)]: "Ste" in the beginning of the reactor name indicates the sterilized reactors.

Figure 2. Experimental set-up of the gas production rate experiment, Phase I: 5-L glass bottles with anaerobic headspace connected to 2-L aluminium gas bags, incubated at room temperature.

Bottles containing only inoculum and water were used as blanks. The CH_4 production from the blanks was subtracted from the CH_4 production of waste samples. Mesophilic inoculum was the same as the inoculum used in previous experiments (from VA Syd Sjölunda WWTP), which was used for incubation

at room temperature and 37 °C. Thermophilic inoculum, which was used for incubation at 55 °C, was collected from the Snertinge biogas plant (DK). The biogas volume was measured using a water displacement method. CH_4 content of the produced biogas was measured using a gas chromatograph (490-PRO Micro GC).

The FOD kinetic constant (yr^{-1}) was calculated by the following equation:

$$k = \frac{-\ln\left(\frac{m_t}{m_0}\right)}{t - t_{lag}} \quad \text{(Eq. 2)}$$

where m_t and m_0 are the masses of organic carbon (g) at time t and 0, respectively, while t and t_{lag} are the degradation time and lag-phase time (yr), respectively. The ratio between m_t and m_0 was found by the following equation:

$$\frac{m_t}{m_0} = \frac{\text{Ultimate } CH_4 \text{ Potential } (t_\infty) - \text{Cumulative } CH_4 \text{ (t)}}{\text{Ultimate } CH_4 \text{ Potential } (t_\infty)} \quad \text{(Eq. 3)}$$

where Ultimate CH_4 Potential (t_∞) is the theoretical CH_4 potential from each reactor, calculated by multiplying the BMP of each sample (g CH_4/kg waste) by the mass of waste sample in the reactor (kg), and Cumulative CH_4 (t) is the CH_4 production (g) from each incubation bottle. The details of the calculations can be found in Section 2.4.2 of Paper **I**.

2.3 Determination of gas recovery efficiency at the landfills

GRE can be calculated by dividing the CH_4 recovery rate by the total CH_4 production rate. In order to calculate the total CH_4 production at the landfills, five terms of the CH_4 mass balance (Eq. 1) must be quantified: CH_4 recovery, CH_4 emission, CH_4 oxidation, CH_4 migration and changes in CH_4 storage. The following sections describe how these terms were quantified.

2.3.1 Methane recovery measurements

The LFG flow rates and the CH_4 content of the recovered LFG were supplied by the landfill operators. These data were recorded every 2 min at normal temperature and pressure (NTP; T = 293.15 K, P = 1 atm). We calculated the CH_4 recovery rates (Nm^3 CH_4/h) by multiplying the LFG flow rates (Nm^3 LFG/h) by their corresponding CH_4 concentration (v/v %). Then, the CH_4 volumetric recovery rate (Nm^3 CH_4/h) was converted to mass (kg CH_4/h) by using the density of CH_4 at NTP (0.668 kg/m^3). Finally, an average CH_4

recovery rate was calculated for the time when emission measurements were performed.

2.3.2 Methane emission measurements

The tracer gas dispersion method was used in this study for the quantification of total CH_4 emissions from both sites. In this method, CH_4 emissions can be quantified by performing several traverses downwind, perpendicular to the plume from the landfill by a vehicle, which carries the analytical instrument, in order to measure the atmospheric concentration of CH_4 and tracer gas. Figure 3 illustrates the measurement concept.

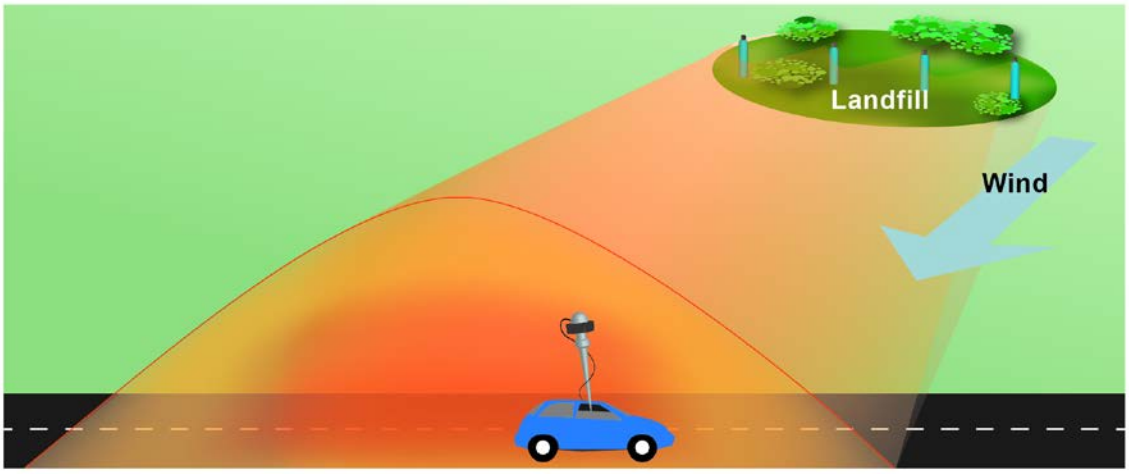

Figure 3. Illustration of the tracer gas dispersion method: an analytical instrument mounted on a vehicle measures the atmospheric concentration of CH_4 and tracer gas downwind of the landfill (Mønster et al., 2014).

The CH_4 emission rate (E_{CH_4}) can be calculated by the following equation (Mønster et al., 2015, 2014):

$$E_{CH_4} = Q_{tracer} \times \frac{\int_{plume\ start}^{plume\ end} C_{CH_4}\, dx}{\int_{plume\ start}^{plume\ end} C_{tracer}\, dx} \times \frac{MW_{CH_4}}{MW_{tracer}} \qquad \text{(Eq. 4)}$$

where Q_{tracer} is the release rate of tracer gas (kg/h), C_{CH_4} and C_{tracer} are the concentrations of CH_4 and tracer gas downwind (ppbv) above the background, x is the distance across the plume (m), and MW_{CH_4} and MW_{tracer} are the molar weight of CH_4 and tracer gas, respectively.

In total, six campaigns were performed from January to October 2016. Each campaign started with a screening of the landfills to find CH_4 emission hotspots, where tracer bottles were placed. Acetylene was used as tracer gas.

Gas concentrations were measured by a $C_2H_2/CH_4/H_2O$ analyser (G2203, Picarro, Inc., Santa Clara, CA), based on cavity ring-down spectroscopy.

Wind speed, wind direction, temperature and barometric pressure recorded during the time interval of the measurements are shown in Table 1 of Paper **II**. In general, measurements were performed on days with relatively stable weather conditions, as these parameters can impact CH_4 emissions from landfills (Christophersen et al., 2001; Czepiel et al., 2003; Xu et al., 2014).

2.3.3 Methane oxidation measurements

Stable carbon isotopic analysis was used to measure CH_4 oxidation occurring at the top layer of the landfills. The methanotrophs oxidize ^{12}C slightly faster than ^{13}C (Chanton et al., 1999). Thus, when CH_4 passes through the top cover and oxidation occurs, it becomes more ^{13}C-enriched. An analysis of the carbon isotopic composition, $\delta^{13}C$, shows how ^{13}C-enriched the sample is. The CH_4 oxidation efficiency (f_{ox}) can then be calculated using the following equation (Börjesson et al., 2001; Chanton et al., 1999):

$$f_{ox} = \frac{\delta_{excess} - \delta_A}{1000\ (\alpha_{ox} - \alpha_{trans})} \quad \text{(Eq. 5)}$$

where δ_{excess} is the excess of $\delta^{13}C$ in the downwind plume or surface air corrected for background samples, δ_A is the $\delta^{13}C$ of anoxic CH_4, α_{ox} is fractionation factor associated with landfill cover, and α_{trans} is the transport fractionation factor, which was assumed to be 1 (Börjesson et al., 2007).

The α_{ox} was determined by the incubation of 200 g of soil samples from the top layer of the landfills in 1-L sealed glass bottles at room temperature. Three top-layer samples were taken: two from Stige Ø (northern and southern sections) and one from the SW cell of Odense Nord. CH_4 was added to the incubation bottles when the experiment was initiated to reach concentrations of 10-11% CH_4 (C_0). Gas samples were taken over time for the determination of CH_4 concentration (C) and isotopic analysis ($\delta^{13}C_t$) until the CH_4 concentration was lower than 1%. The α_{ox} was determined by plotting $\delta^{13}C_t$ against $\ln\left(\frac{C}{C_0}\right)$, and using the following equation (Coleman et al., 1981):

$$\delta^{13}C_t \cong 1000\left(\frac{1}{\alpha_{ox}} - 1\right)\ln\left(\frac{C}{C_0}\right) + \delta^{13}C_{t=0} \quad \text{(Eq. 6)}$$

When $\delta^{13}C$ is plotted versus $\ln(\frac{C}{C_0})$, the slope of the line fitted to the data is $1000\left(\frac{1}{\alpha_{ox}} - 1\right)$.

By determining the CH_4 oxidation efficiency (f_{ox}), it is possible to calculate the CH_4 oxidation rate (MO; kg/h) as a function of the total CH_4 emission rate (E; kg/h) using the following equation (Börjesson et al., 2007):

$$MO = f_{ox}\left(\frac{E}{1-f_{ox}}\right) \quad \text{(Eq. 7)}$$

Anoxic zone gas samples were taken from four MPR modules at Stige Ø and the MPR module of the SW cell of Odense Nord. The downwind samples were taken from the downwind plume and also the surface air across the Stige Ø landfill and the SW cell of the Odense Nord landfill. The background gas samples were taken upwind of the Stige Ø landfill and the SW cell of the Odense Nord landfill. Figure 4 shows the locations of the collected anoxic zone, upwind and downwind gas samples, surface air samples, and soil samples for the determination of α_{ox}. Gas samples for CH_4 oxidation measurements were taken in May 2016 and February 2017. Soil samples were taken in May 2016.

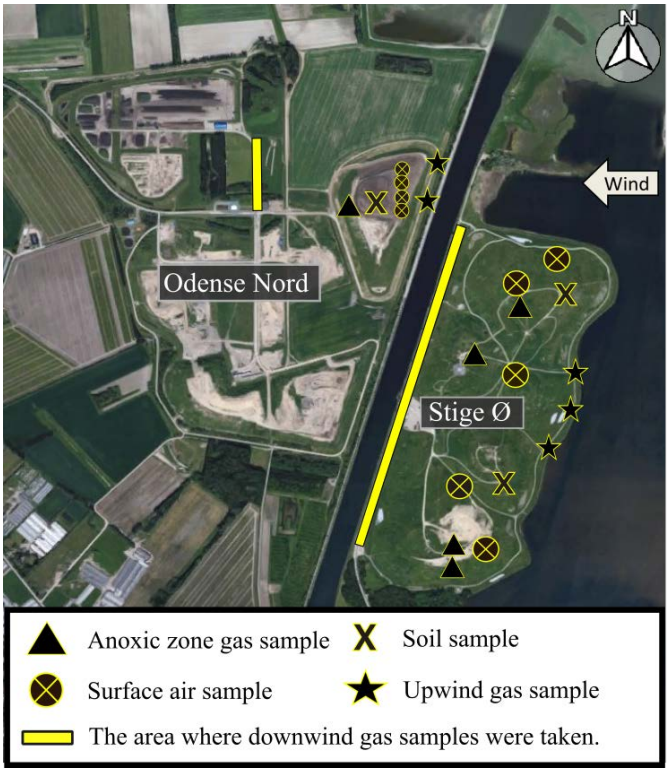

Figure 4. Locations of the collected anoxic zone, upwind and downwind gas samples, surface air samples, and soil samples. As the number of the downwind gas samples and their locations were slightly different during the two sampling days in May 2016 and February 2017, only the area of the sampling is shown, rather than the exact sample locations (Imagery ©2016 Google, Aerodata International Surveys).

2.3.4 Methane lateral migration

There is a drainage trench along the edge of the Stige Ø landfill to collect leachate and surface water. CH_4 migrating laterally off-site will be emitted to the atmosphere from the same trench surrounding the landfill, and thus is accounted for in CH_4 emission measurements. Therefore, the CH_4 migration from the Stige Ø landfill was assumed to be zero.

The bottom lining system in the SW and mixed waste cell of the Odense Nord landfill is composed of a HDPE membrane on top of a clay layer. An estimation of the CH_4 migration from the Odense Nord landfill was calculated by multiplying the area of the SW and mixed waste cells (19.1 hectares) by 4.2×10^{-7} kg $CH_4/(m^2 . d)$, which can pass through an HDPE liner with a thickness of 1.5 mm (Pauly, 1989; Lim, 1995). It should be noted that the resistance of the clay layer was not considered in this study.

2.3.5 Changes in methane storage

This term could be calculated by shutting down the gas recovery system for a period of time to let the gas build up in the waste body of the landfill and then monitor the CH_4 recovery rate before and after system shutdown. The amount of additional CH_4 recovered after shutdown, in comparison to prior shutdown, is an approximation of $\Delta CH_4\ storage$. However, it was not possible to determine this term in the current study because shutting down the gas recovery system could pose a risk to the health of citizens using the recreational centre on the Stige Ø landfill. Thus, $\Delta CH_4\ storage$ was assumed to be zero.

2.3.6 Methane production modelling

The total CH_4 production rate in these landfills was estimated using the Afvalzorg model and compared with the results of field measurements. The Afvalzorg model was developed by a Dutch waste management company and accommodates up to eight waste categories. The annual amount of deposited waste was supplied by the landfill operators. This inventory consisted of up to 31 waste categories.

These waste categories were fitted into the eight waste categories of the Afvalzorg model, using the guideline developed by Scheutz et al. (2007). This model was chosen on the basis that a previous study (Mou et al., 2015a), which compared the results of LandGEM, IPCC and Afvalzorg models with the measured CH_4 emissions, found the latter to be the most suitable for the estimation of CH_4 production at Danish landfills.

In addition to modelling the gas production with the default value of the Afvalzorg model, we modelled gas production using a revised version. In the revised version, site-specific BMPs and k-values of SW, sludge and bulky mixed waste, measured by Mou et al. (2015b, 2014), were used instead of the default values of the model. The details of the revised model can be found in Section 2.6 of Paper **II**.

2.4 Impact of meteorological parameters on methane extraction from the landfills

2.4.1 Data collection

The LFG flow rates and the CH_4 content of the recovered LFG were supplied by the landfill operators. These data were recorded every 2 min at NTP (T = 293.15 K, P = 1 atm). The CH_4 flow rates (Nm^3 /h) were calculated by multiplying the LFG flow rates (Nm^3 LFG/h) by their corresponding CH_4 concentration (v/v %). Next, we converted these data to hourly averages.

The meteorological parameters (barometric pressure, ambient temperature, wind speed and solar radiation) were recorded hourly at a weather station at the Odense Nord landfill. One period in 2015 (11.08.2015-06.09.2015) and three periods in 2016 (15.08.2016-25.08.2016, 05.09.2016-11.09.2016 and 05.12.2016-08.12.2016) were chosen for this study, given that, during these periods, no manual changes were made to the gas recovery system by the landfill operators.

The effect of changes in barometric pressure on the recovered LFG was studied by diving the periods into sub-periods, based on decreasing and increasing pressure tendencies, then calculating the changes in barometric pressure and the corresponding LFG data for each sub-period and performing correlation tests on the calculated changes.

2.4.2 Statistical analysis

The Spearman method (Reimann et al., 2008) was used to study the correlation between the parameters. This method gives a correlation coefficient (r) between -1 and +1, which shows how strongly the two variables are correlated. Correlation coefficients of ±1 show a perfect correlation between the variable, while 0 shows that there is no correlation.

In addition, p-values were calculated to address whether the correlation coefficients were significantly different from 0. In general, when $p \geq 0.10$, we

considered that the correlation was not statistically significant (Reimann et al., 2008). The details about the significance of the p-values can be found in Section 2.3 of Paper **III**.

3 Results and discussion

3.1 Methane production from shredder waste at landfills

3.1.1 Waste characterization

The TS, VS, TC, TOC and BioC of the waste samples were 82-91%, 15-32%, 12-24%, 11-21% and 4.2-8.3%, respectively. In general, the TS, VS, TC, TOC content of the waste samples were comparable to the characterization results of a previous study on SW from three Danish landfills (Mou et al., 2014), except for FUS samples, which had higher TC and TOC contents (24% and 21%, respectively) in comparison to the TC and TOC contents of SW (12-13%, and 10-11%, respectively) reported by Mou et al. (2014).

The high TS content indicated that the waste samples were very dry. FUS had the highest VS, TC, TOC and BioC. The reason is that the FUS sample was a fresh sample, which had been recently deposited in the landfill and not undergone anaerobic digestion in the landfill, and thus had the highest amount of volatile compounds and carbon.

The old samples (SW2009 and SW2012) had lower VS, TC, TOC and BioC in comparison to the fresh sample (FUS), which is most likely due to anaerobic digestion following their deposition in the landfill. FS had the lowest VS, TOC and BioC. This was expected because the fine fraction had a higher amount of mineral and inert material (Ahmed et al., 2014). The BioC to TOC ratios were 39-53%, indicating that a high portion of the organic carbon (47-61%) originated from fossil sources.

3.1.2 Biochemical methane potential

Figure 5 shows the cumulative CH_4 production of SW samples during the BMP experiment. FUS had the highest CH_4 production, as it was the fresh sample and had not undergone anaerobic digestion in the landfill. The BMP of the control was 421 mL CH_4/g VS, which was in the accepted range of 315-439 mL CH_4/g VS (Hansen et al., 2004). FS samples produced lower CH_4 than FUS samples, as the fine fraction contained more mineral and inert material (Ahmed et al., 2014).

The BMP of samples and the CH_4 recovery rates were 0-6.2 kg CH_4/ton waste and 0-4.5%, respectively. The SW2009 and SW2009+Fe produced

lower CH_4 than the blanks; thus, their BMPs and CH_4 recovery rates were considered as 0. The measured BMPs and CH_4 recovery rates were comparable to the BMPs and CH_4 recovery rates of 6.2-9.1 kg CH_4/ton waste and 4.4-6.4%, respectively, for SW from three Danish landfills, measured by Mou et al. (2014).

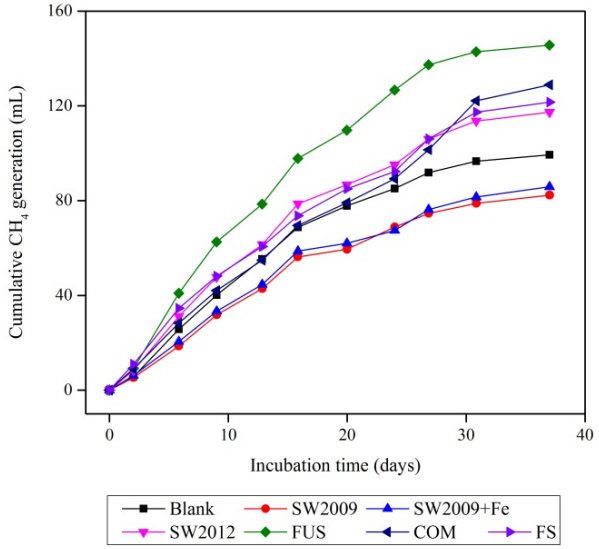

Figure 5. Cumulative CH_4 production of different SW samples in the BMP experiment.

3.1.3 Biocorrosion experiment

The cumulative H_2 and CH_4 production curves are presented in Figure 2 of Paper **I**. Fe, Al and Zn produced H_2 when in contact with water, while Cu did not. Production of H_2 was not observed in the bottles containing inoculum. This is most likely because hydrogenotrophic methanogenesis is a fast process, and the produced H_2 is utilized rapidly for CH_4 production (Gerardi, 2003).

The addition of Fe, Al and Zn to inoculum resulted in higher CH_4 production than inoculum alone, while the addition of Cu resulted in lower CH_4 production. Fe, Al and Zn have been reported previously to increase CH_4 production (Belay and Daniels, 1990; Hu et al., 2015; Lorowitz et al., 1992), while Cu has been reported to inhibit CH_4 production (Ahring and Westermann, 1985; Jin et al., 1998). Overall, this experiment showed that H_2 can be produced by the anaerobic corrosion of Fe, Al and Zn, while the produced H_2 can be consumed by hydrogenotrophic methanogens to convert CO_2 into CH_4.

3.1.4 Gas production rate experiment

Figure 6 shows the CH_4 production of SW samples during two experimental phases of the gas production rate experiment at three different temperatures. FUS, which had the highest BMP, produced the highest amount of CH_4 in this experiment. Sterilized reactors did not produce CH_4. This showed that CH_4 production from SW was only biological.

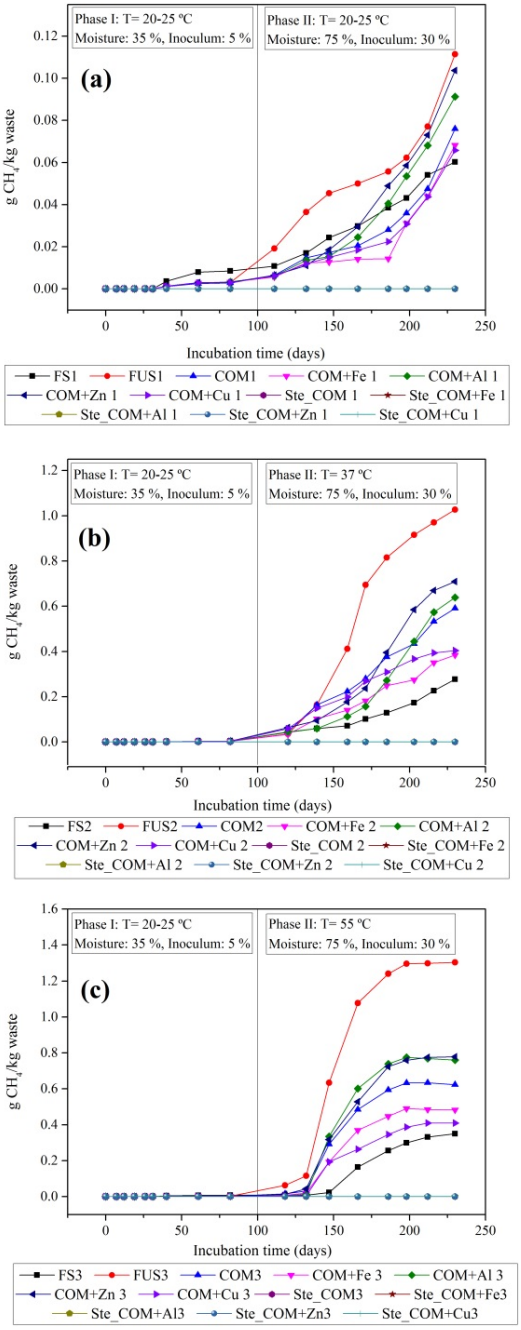

Figure 6. Cumulative CH_4 production from SW and sterilized SW in the gas production rate experiment.

The addition of Al and Zn to COM samples (COM+Al and COM+Zn in Figure 6) resulted in higher CH_4 production compared to COM alone, while adding Fe and Cu resulted in lower CH_4 production. Moreover, the COM+Al and COM+Zn reactors had lower headspace CO_2 and higher CH_4 concentrations in comparison to COM (Paper **I**, Table 8). This could mean that a portion of existing CO_2 in the produced biogas reacted with the H_2 produced by corrosion of these metals and led to higher CH_4 concentrations.

When comparing the two phases (Phase I and Phase II) in Figure 6a, it can be observed that the higher moisture and inoculum content resulted in significantly higher CH_4 production from SW. Higher CH_4 production as a result of increasing moisture and inoculum content has been reported previously (Forster-Carneiro et al., 2007; Fujishima et al., 2000; Liotta et al., 2014; Lopes et al., 2004). It can be seen from Phase II in Figure 6 that higher temperature resulted in higher CH_4 production, when moisture and inoculum content was the same in all reactors. These results emphasize the importance of moisture and temperature in CH_4 production from SW in landfills.

The calculated k-values were higher at 55 °C (0.235 to 0.488/yr) in comparison to 37 °C (0.220 to 0.429/yr) and room temperature (0.033 to 0.075/yr). The details of the calculation can be found in Table 9 of Paper **I**. This is reasonable, as a higher temperature can result in faster anaerobic digestion and thus a higher k-value. The calculated k-values at room temperature were higher than the calculated k-values of 0.016-0.017/yr, as measured by Mou et al. (2015b) at room temperature for SW from three Danish landfills, which is most likely due to higher moisture content and inoculum addition in this study.

3.2 Determination of gas recovery efficiency at the landfills

3.2.1 Methane emission measurements

Overall, six CH_4 emission measurement campaigns were performed. Figure 7 shows CH_4 and tracer gas plumes measured downwind of the landfills on October 7, as an example. The measured plumes in the rest of the campaigns can be found in Figure 3 of Paper **II**. The screening results showed elevated CH_4 concentrations (up to 16 ppm) at four locations: the mixed waste cell, shredder waste cell and composting facility at the Odense Nord site, and next to the drainage trench around the Stige Ø landfill, especially in the northern section of the landfill. Thus, four tracer bottles were placed in these four

locations, which are marked with orange triangles in Figure 7. The screening results are shown in Paper **II**.

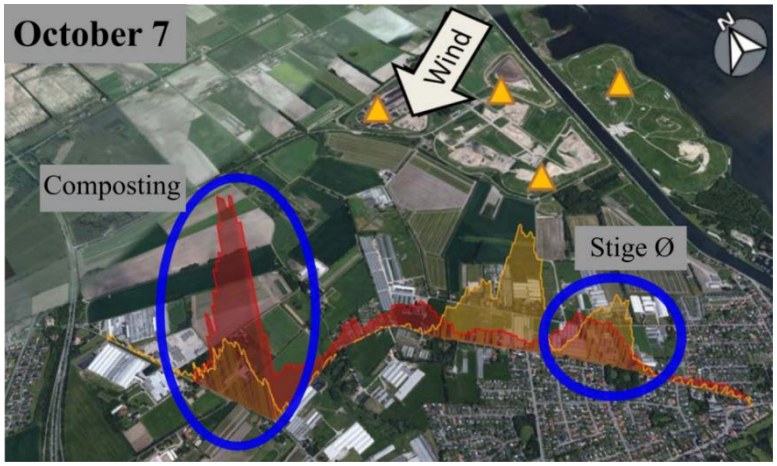

Figure 7. CH_4 (red) and tracer gas (yellow) plumes measured downwind of the landfills.

The total CH_4 emissions from the two landfills combined ranged between 29.1 and 49.6 kg/h. The emissions from the southern part of Stige Ø and the mixed waste cell of Odense Nord were 4.3-5.4 kg CH_4/h, when it was possible to distinguish them. This showed that the majority of CH_4 emissions come from the other parts of the landfills, namely, the northern part of Stige Ø and the shredder waste cell of Odense Nord. The CH_4 emissions in September and October 2016 (29.1-33.5 kg/h) were lower than the emissions in January and March 2016 (49.6 and 45 kg/h, respectively), most probably because of the commencement of gas recovery from the second section of the SW cell in May 2016.

The measured CH_4 emissions in this study were higher than most of the measured CH_4 emissions from Danish landfills using the tracer gas dispersion method by Mønster et al. (2015) and Scheutz et al. (2011b, 2011c). This was probably due to a higher amount of deposited waste in the Stige Ø and Odense Nord landfills, in comparison to the other studied Danish landfills.

The measured CH_4 emissions in this study were comparable to the lowest CH_4 emissions from Swedish landfills measured by Börjesson et al. (2009). These landfills with lower CH_4 emissions (Kristianstad, Visby and Sundsvall landfills) received lower organic waste in comparison to the rest of the landfills in the study (Börjesson et al., 2009). The measured CH_4 emissions in this study were significantly lower than CH_4 emissions from US landfills measured by Mosher et al. (1999) and most of the Swedish landfill measured

by Börjesson et al. (2009), most likely due to the ban on the landfilling of organic waste in Denmark since 1997.

3.2.2 Methane oxidation measurements

The measured fractionation factor (α_{ox}) for the northern and southern parts of the Stige Ø samples and the SW sample were 1.015, 1.024 and 1.025, respectively. The measured fractionation factors in this study are comparable to the measured fractionation factors in previous studies (Börjesson et al., 2001; Chanton and Liptay, 2000; Chanton et al., 2008; Liptay et al., 1998). The results of stable carbon isotopic analysis performed on downwind, upwind and anoxic zone samples, along with the calculated CH_4 oxidation efficiencies (f_{ox}), are shown in Table 2.

Table 2. The carbon isotopic ratio of CH_4 in downwind, upwind and anoxic zone samples from the Odense Nord and Stige Ø landfills and the calculated CH_4 oxidation efficiency.

Landfill	Date	(P)/ (S) [(a)]	DW [(b)] CH_4 conc.[(c)]	DW [(b)] $\delta^{13}CH_4$	UW [(d)] CH_4 conc. [(c)]	UW [(c)] $\delta^{13}CH_4$	δ_{excess}	Anoxic CH_4; δ_A	f_{ox} (%)
Odense Nord	February 2017	P	2.14	-50.20	2.02	-49.43	-61.67	-69.61	32
	May 2016	P	2.48	-49.97	1.97	-47.99	-57.62	-66.80	37
	May 2016	S	24.23	-59.94	1.97	-47.99	-62.51	-66.80	17
Stige Ø	February 2017	P	2.34	-49.66	2.03	-48.90	-55.96	-57.21	6
	May 2016	P	2.74	-48.55	1.92	-46.68	-53.10	-54.38	7
	May 2016	S	3.98	-48.45	1.92	-46.68	-52.65	-54.38	9

[(a)]: This column shows whether the downwind samples were taken in the downwind plume (P) or on the surface (S) across the landfill.
[(b)]: DW = downwind
[(c)]: conc. = concentration
[(d)]: UW = upwind

Higher CH_4 concentrations were observed in the surface gas samples in comparison to the plume downwind samples. This is reasonable, as the downwind plume samples were more diluted in comparison to the surface samples. The $\delta^{13}CH_4$ values of the downwind and upwind samples (Table 2) were comparable to those reported in previous studies (Börjesson et al., 2007; Scheutz et al., 2011a).

Table 3 presents an overview of reported oxidation efficiencies in the literature. A wide range of oxidation efficiencies (0-94%) is observed in previous studies. The calculated CH_4 oxidation efficiency at the Odense Nord landfill (17-37%) was comparable to the calculated CH_4 oxidation efficiency of 16-41% at the Fakse landfill in Denmark using stable carbon isotopic

analysis (Scheutz et al., 2011a). The calculated CH_4 oxidation efficiency at the Stige Ø landfill (6-9%) was similar to the measured CH_4 oxidation efficiency of 6-7% at the Metro and Clearview landfills in the US (Chanton et al., 2011). The average CH_4 oxidation efficiency of both landfills was 18%, which was used to calculate CH_4 oxidation rate (kg/h) using Eq. 7.

Table 3. Overview of reported oxidation efficiencies in the literature.

Reference	Oxidation efficiency (%)	Methodology
Börjesson et al. (2001)	0-94	Determined by stable carbon isotopic analysis at two closed Swedish landfills in summer and winter.
Börjesson et al. (2007)	5-42	Determined by stable carbon isotopic analysis at four active and two closed Swedish landfills.
Chanton and Liptay (2000)	20	Average oxidation efficiency determined by stable carbon isotopic analysis in different seasons at a US landfill with mulch, topsoil and clay as the top layer.
Chanton et al. (1999)	0-24	Determined by stable carbon isotopic analysis at two US landfills with active and closed sections and different types of top cover.
Chanton et al. (2009)	36	Average oxidation efficiency determined by review of 42 determination of oxidation efficiency using both column measurements and field measurements in a variety of landfill top covers.
Chanton et al. (2011)	36-38	Average oxidation efficiency determined by stable carbon isotopic analysis at 20 US landfills with intermediate covers.
Scheutz et al. (2011a)	16-41	Determined by stable carbon isotopic analysis at a Danish landfill, where sandy clay loam was the final top cover, while the temporary top cover varied from sandy clay loam to a more porous sandy loam with a high content of wood and plant material.

3.2.3 Total methane production rate and gas recovery efficiency

The measured CH_4 emission, recovery and oxidation rates, as well as the calculated total CH_4 production rate and the GRE, are shown in Table 4. The calculated lateral CH_4 migration was 0.003 kg/h, which was negligible in comparison to the CH_4 emission, recovery and oxidation rates. Very low lateral CH_4 migration was expected as a geomembrane (HDPE) bottom liner is in place.

The CH_4 recovery rate was 84.8-115.3 kg/h. The higher CH_4 recovery rate in September and October 2016 (108.1-115.3 kg/h), in comparison to January and March 2016 (84.8-88.1 kg/h), was because of the initiation of gas recovery from the second section of the SW cell in May 2016. The average total CH_4 production from both landfills combined was 147.4 kg/h. Small variations (CV = 0.02) were observed between the obtained total CH_4 production rates.

Table 4. CH_4 emission, oxidation, recovery and GRE of the landfills.

Date	E = CH_4 emission rate (kg/h)	R = CH_4 recovery rate (kg/h)	MO = CH_4 oxidation rate (kg/h)	Total CH_4 production rate (E+R+MO; kg/h)	Gas recovery efficiency (%)
January 14, 2016	49.6	88.1	10.9	148.6	59
March 15, 2016	45.8	84.8	10.1	140.7	60
September 8, 2016	29.1	115.3	6.4	150.8	76
October 6, 2016	31.2	109.2	6.8	147.2	74
October 7, 2016	33.5	108.1	7.4	149.0	73
October 21, 2016	33.0	108.1	7.2	148.3	73
Average	37.0	102.3	8.1	147.4	69
Standard deviation	8.5	12.6	1.9	3.5	7
Coefficient of variation (CV) [(a)]	0.23	0.12	0.23	0.02	0.11

[(a)]: CV = standard deviation/average

The calculated GRE was 59-76%. Table 5 presents an overview of previous studies that have determined the GRE by field measurements. Overall, there are relatively few studies in the literature on this topic. A wide range of GRE (14-94%) was observed in previous studies. The measured GRE in this study was comparable to the GRE of 61-78% measured by Börjesson et al. (2009) at the Filborna landfill in Sweden and that of 69-79% measured by Lohila et al. (2007) at the Ämmässuo landfill in Finland, while it was higher than the measured GRE of 29-41% by Börjesson et al. (2009) at the Högbytorp landfill in Sweden and lower than that of 84-88% measured by Spokas et al. (2006) at the Grand Landes landfill in France.

Table 5. Overview of the reported GRE in the literature.

Reference	GRE (%)	Landfill(s) and the type of top cover
Börjesson et al. (2009)	14-78	Seven active and closed Swedish landfills with different types of top cover: clay, mixture of sewage sludge and soil and mixture of wood chips and sludge
Lohila et al. (2007)	69-79	An active Finnish landfill with a temporary soil top cover, and compost soil on top of a diamicton and clay layer on the slopes.
Mosher et al. (1999)	90	A US closed landfill with a geomembrane and soil top cover
Mønster et al. (2015)	41-81	Five active and closed Danish landfills; no information about the type of their top cover
Spokas et al. (2006)	41-94	Three active and closed French landfills with different types of top cover: clay cover, geosynthetic clay liner and geomembrane cover

3.2.4 Modelling and comparison with field measurements

Figure 8 shows the annual CH_4 production rate at the Odense Nord and Stige Ø landfills, estimated by a default and a revised version of the Afvalzorg model. The estimated CH_4 production rate by the revised version of the model was lower than the default version because of the lower BMPs and k-values in the revised version.

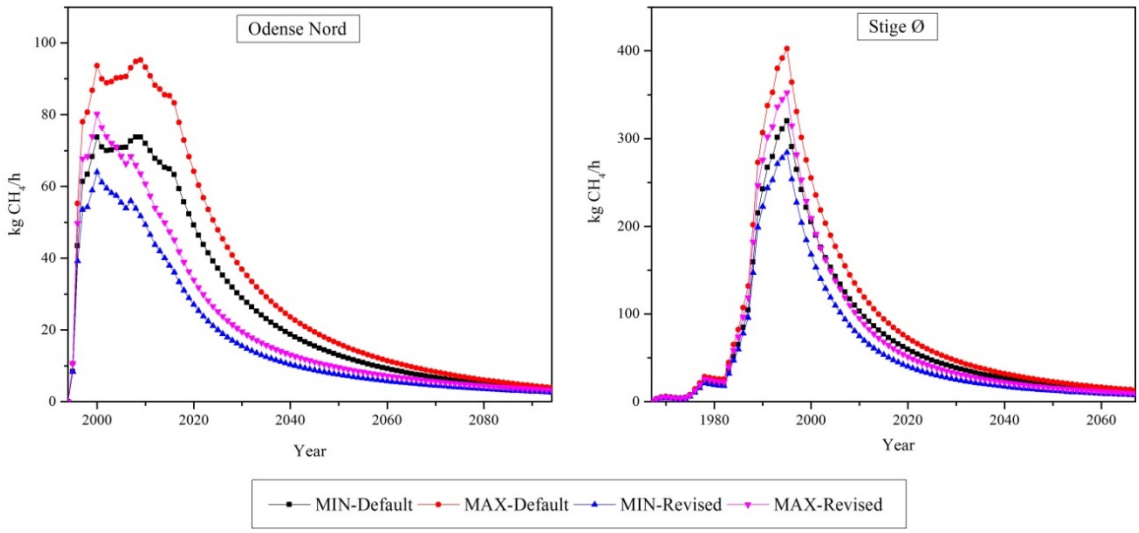

Figure 8. Minimum and maximum annual CH_4 production rate at the Odense Nord (left) and Stige Ø (right) landfills estimated by Afvalzorg model using default and revised BMPs and k-values.

The total CH_4 production rate in 2016 for both landfills combined, as estimated using default and revised Afvalzorg models, was 136.3-172.6 kg/h and 86.3-109.3 kg/h, respectively. This indicated a good agreement between the average CH_4 production obtained by field measurements (147.4 kg CH_4/h) and the average estimated CH_4 production by the default Afvalzorg model in 2016 (154.4 kg CH_4/h).

This is surprising, as previous studies, which compared modelling and experimental results (Mou et al., 2015b; Scheutz et al., 2011b), showed a significant overestimation or underestimation by FOD models. This could be related to the uncertainties associated with waste sampling and field measurements, and to the value of different parameters used in the model. For instance, Börjesson et al. (2009) observed a good agreement between field measurements and the results of the IPCC model, when the fraction of degradable carbon, when dissimilated, was set to 0.54.

3.3 Impact of meteorological parameters on methane extraction from the landfills

Table 6 shows the results of correlation tests between meteorological parameters and LFG data. The highest correlation coefficients were observed between barometric pressure and LFG data. Each parameter is discussed individually in the following sections.

Table 6. Correlation coefficients and p-values between meteorological parameters and LFG data during the periods studied in 2015 and 2016.

Year	Parameters	n [(a)]	LFG CH_4 concentration (%)	LFG flow (Nm^3/h)	CH_4 flow (Nm^3/h)
2015	Barometric pressure (mbar)	600	-0.73 (***)	0.51 (***)	-0.37 (***)
	Changes in barometric pressure (mbar)	13	-0.93 (***)	0.85 (***)	-0.80 (***)
	Ambient temperature (°C)	600	-0.12 (+)	0.27 (***)	0.19 (***)
	Wind speed (m/s)	600	0.15 (***)	-0.22 (***)	-0.02 (+)
	Solar radiation (W/m^2)	600	-0.07 (*)	0.03 (+)	-0.04 (+)
2016	Barometric pressure (mbar)	444	-0.56 (***)	0.64 (***)	0.01 (+)
	Changes in barometric pressure (mbar)	13	-0.81 (**)	0.89 (***)	0.20 (+)
	Ambient temperature (°C)	444	0.47 (***)	-0.49 (***)	0.27 (***)
	Wind speed (m/s)	444	-0.25 (***)	0.27 (***)	-0.06 (+)
	Solar radiation (W/m^2)	444	0.20 (***)	-0.21 (***)	0.08 (*)

***: $p < 0.001$, very high significance
**: $0.001 \leq p < 0.01$, high significance
*: $0.05 \leq p < 0.10$, weak significance
+: $p \geq 0.10$, no significance
[(a)]: n = number of observations

3.3.1 Barometric pressure and changes in barometric pressure

Barometric pressure correlated strongly and significantly with LFG CH_4 concentration and LFG flow (Table 6). The correlation was negative with LFG CH_4 concentration and positive with LFG flow. A weaker correlation was observed between barometric pressure and CH_4 flow in 2015 ($r = -0.37$), but the correlation was statistically significant. CH_4 flow showed a very weak correlation with barometric pressure in 2016 ($r = 0.01$), which will be discussed later in this chapter.

Figure 9 shows barometric pressure against LFG CH_4 concentration, LFG flow and CH_4 flow in the period 11.08.2015-06.09.2015. It can be seen from the figure that higher barometric pressure corresponds to lower CH_4 concentrations, higher LFG flow and lower CH_4 flow. Czepiel et al. (2003) found a negative correlation between barometric pressure and the CH_4

emission rate at a US landfill. However, to the best of our knowledge, the impact of barometric pressure on LFG recovery has not been studied before.

At higher barometric pressure, the pressure difference, which is the driving force for advective gas emissions, between inside the landfill and the atmosphere decreases. This results in lower advective LFG emissions and thus higher LFG flow recovered by the gas recovery system; meanwhile, the recovered LFG is more diluted because more air has flowed inside the landfill and thus the recovered gas has lower CH_4 concentration.

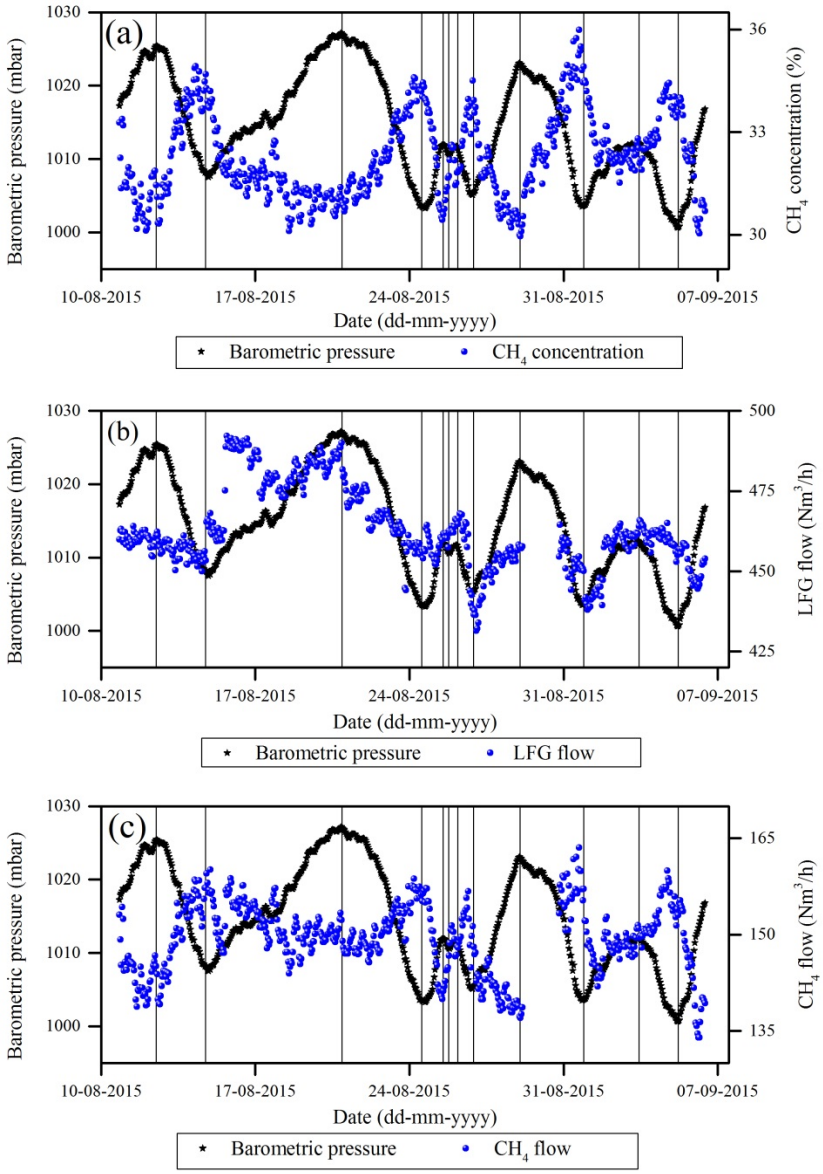

Figure 9. Barometric pressure against LFG CH_4 concentration (a), LFG flow (b) and CH_4 flow (c) in the period 11.08.2015-06.09.2015. The black vertical lines show the division into subperiods based on increasing and decreasing pressure tendencies.

CH_4 flow showed a very weak correlation with barometric pressure in 2016 (r = 0.01). This was because the gas engine had reached its maximum capacity in 2016 and could not burn more gas; thus, it was maintaining a constant CH_4 flow by reducing the suction pressure, when CH_4 concentration was increasing. Therefore, the lack of correlation between barometric pressure and CH_4 flow in 2016 was most likely due to the automatic regulation of the gas engine for maintaining a constant CH_4 flow. Figure 10 shows barometric pressure against LFG data in the period 05.12.2016-08.12.2016. It can be seen from Figure 10c that, even though barometric pressure is changing, CH_4 flow is maintained at a fairly constant level, while the influence of barometric pressure on CH_4 flow in 2015 can be seen in Figure 9c.

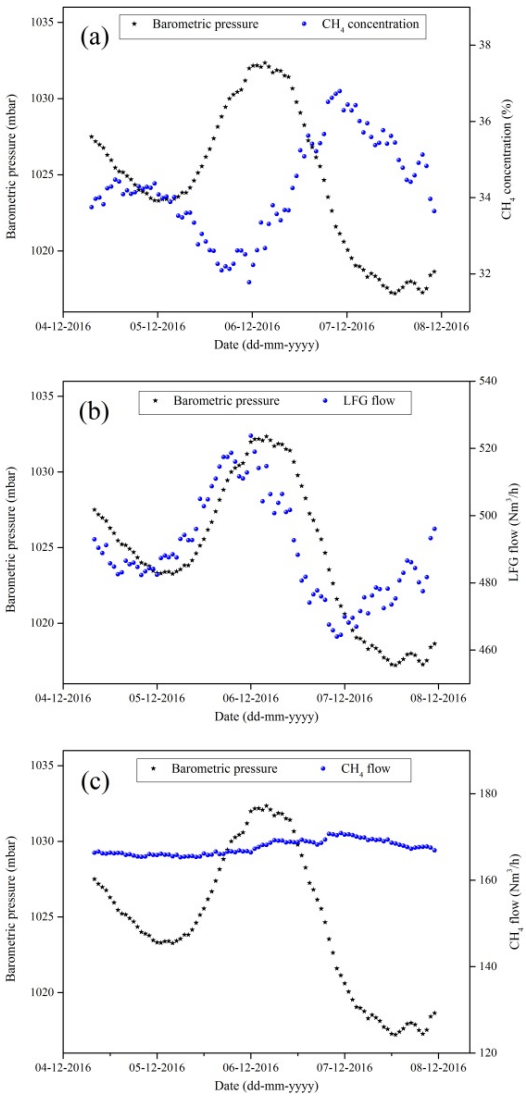

Figure 10. Barometric pressure against LFG CH_4 concentration (a), LFG flow (b) and CH_4 flow (c) in the period 05.12.2016-08.12.2016.

The division of each period into sub-periods (vertical black lines in Figure 9), based on increasing and decreasing pressure tendencies, in order to study the impact of changes in barometric pressure, resulted in 13 sub-periods in 2015 and 13 sub-periods in 2016. Changes in barometric pressure showed strong correlation with changes in CH_4 concentration, LFG flow and CH_4 flow (Table 6).

Correlation of changes in barometric pressure with CH_4 concentration and flow was negative, while the correlation with LFG flow was positive. Figure 11 shows the calculated changes in barometric pressure against changes in CH_4 concentration, LFG flow and CH_4 flow. From the figure, it can be seen that, when barometric pressure decreases, CH_4 concentration and flow increases, while LFG flow decreases.

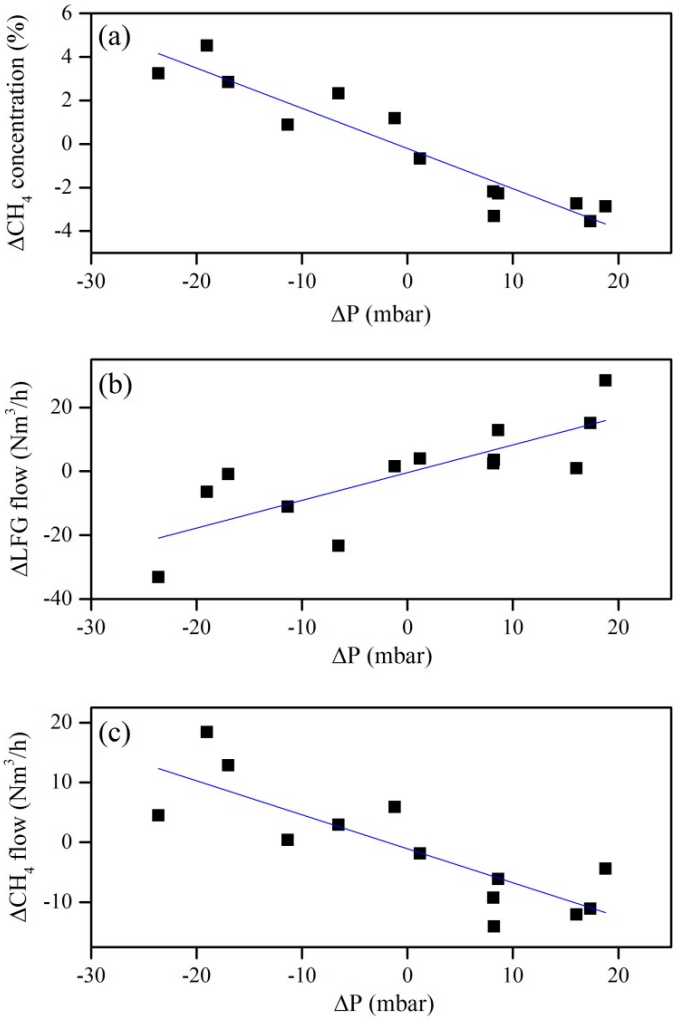

Figure 11. Changes in barometric pressure against changes in LFG CH_4 concentration (a), LFG flow (b) and CH_4 flow (c) in 2015.

Some previous studies (Fredenslund et al., 2010; Gebert and Groengroeft, 2006; Poulsen et al., 2003; Xu et al., 2014) have shown that changes in barometric pressure affect gas emissions from landfills, while Czepiel et al. (2003) found that CH_4 emissions from a landfill in the US were affected by the absolute value of barometric pressure. In our study, LFG recovery was affected by both the absolute value of barometric pressure and changes in barometric pressure. Christophersen et al. (2001) found that LFG emissions from a Danish landfill were affected by both barometric pressure and changes in barometric pressure.

3.3.2 Ambient temperature

LFG data showed a weak correlation coefficient with ambient temperature (Table 6). Figure 12 shows the ambient temperature against LFG CH_4 concentration, LFG flow and CH_4 flow in the period 18.08.2016-25.08.2016. Plots of ambient temperature against LFG data during the rest of the studied periods can be found in Paper **III** and its supplementary material. No visual correlation was observed between ambient temperature and LFG data. This suggests that ambient temperature does not appear to be a controlling factor in the recovered CH_4 concentration of LFG, LFG flow and CH_4 flow.

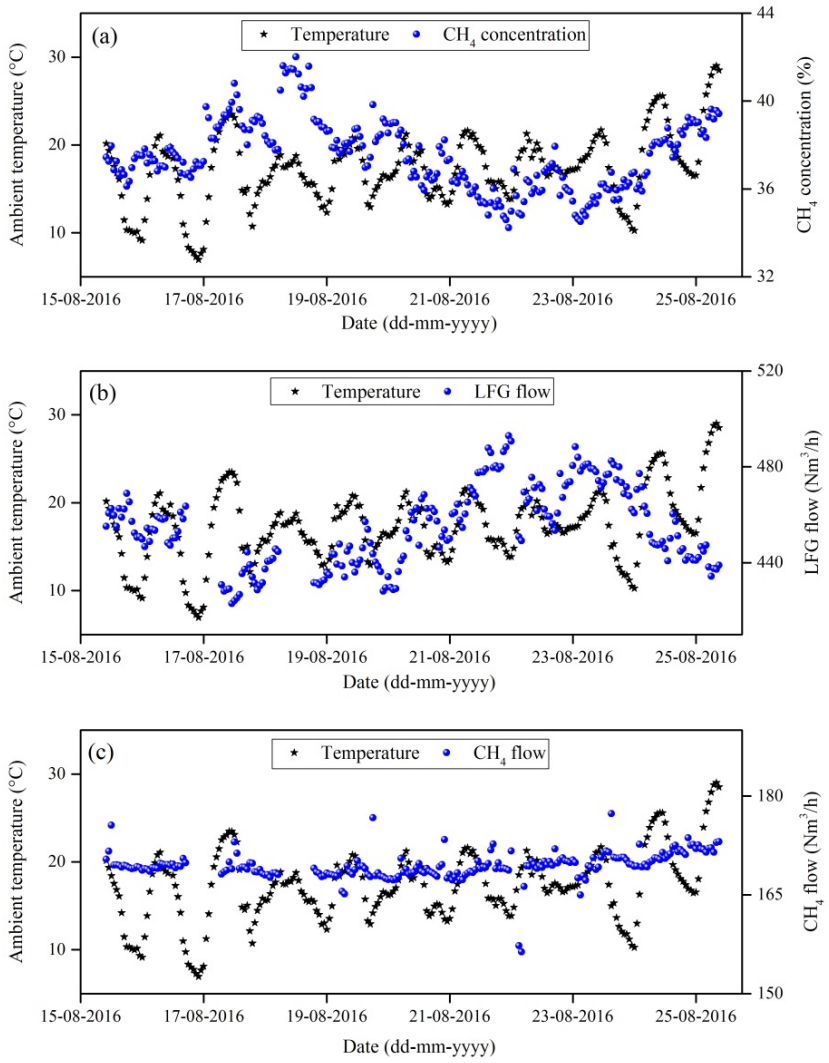

Figure 12. Ambient temperature against LFG CH_4 concentration (a), LFG flow (b) and CH_4 flow (c) in the period 15.08.2016-25.08.2016.

According to the ideal gas law, temperature affects the volume of gases. However, in this study, the LFG recovery data were converted into NTP to avoid this impact. Moreover, temperature can affect CH_4 oxidation (Scheutz et al., 2009) and subsequently CH_4 emissions from landfills (Börjesson and Svensson, 1997), as higher temperature results in higher microbial activities leading to higher CH_4 oxidation and thus lower CH_4 emissions. Moreover, the anaerobic digestion process is temperature-dependent, so temperature can affect the CH_4 production rate (Chen et al., 2008).

The anaerobic digestion of waste in landfills produces heat (Hanson et al., 2010); as the landfills in this study have heights or depths of 20-30 m, the produced heat is most likely maintained inside the body of the landfill. Elevated temperatures inside landfills have been reported in the literature

(Hanson et al., 2010; Olsen and Willumsen, 2013). Therefore, temperatures inside the landfills, which affect the CH_4 production rate, can be different than ambient temperature. This explains why ambient temperature does not affect the recovered LFG flow rate and composition in this study.

3.3.3 Wind speed

The performed correlation tests showed weak correlation between wind speed and LFG data (Table 6). Figure 13 shows wind speed against LFG CH_4 concentration, LFG flow and CH_4 flow in the period 11.08.2015-06.09.2015. No visual correlation was observed between wind speed and LFG data.

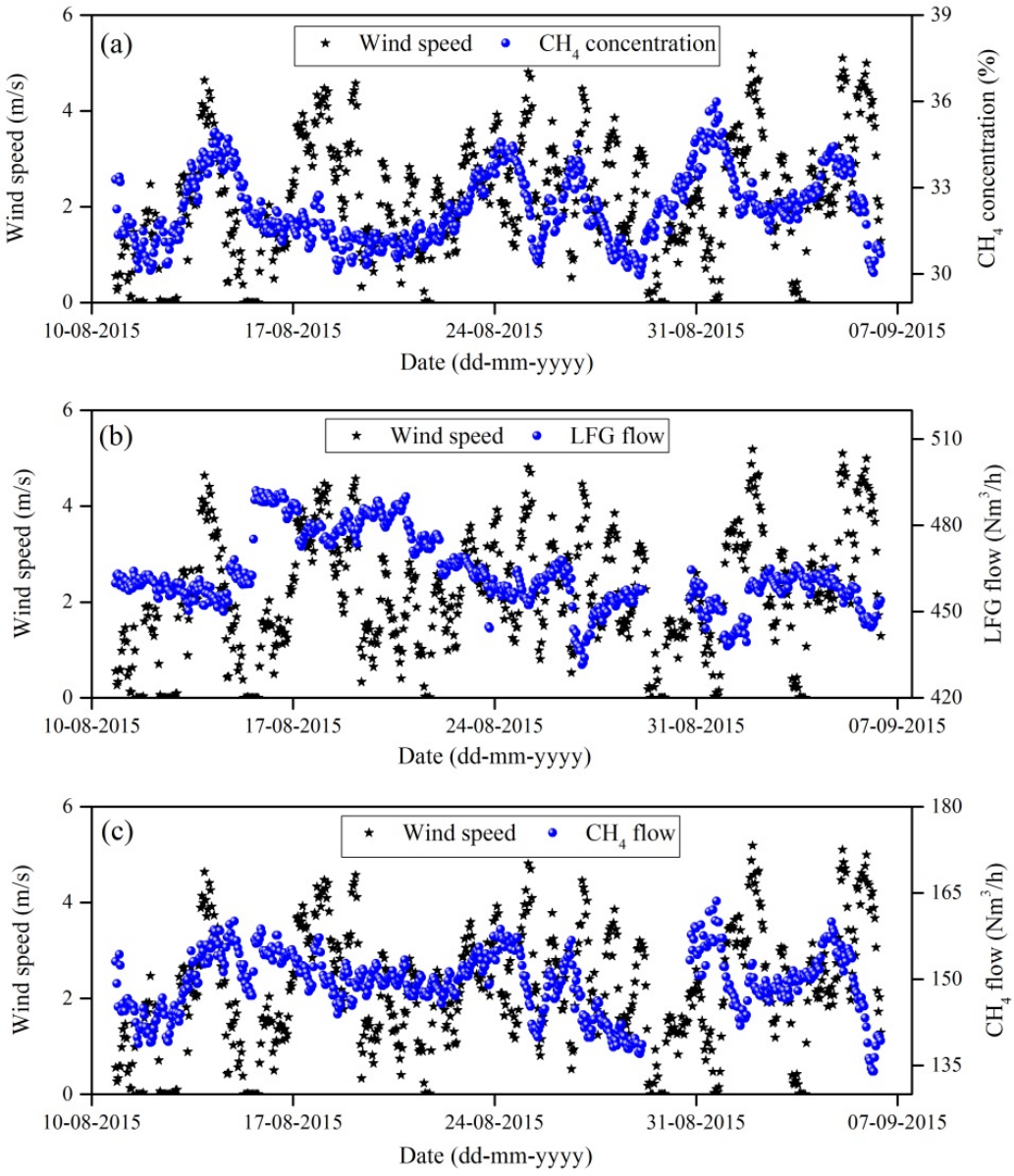

Figure 13. Wind speed against LFG CH_4 concentration (a), LFG flow (b) and CH_4 flow (c) in the period 11.08.2015-06.09.2015.

Table 7 shows the correlation coefficient between wind speed and barometric pressure and LFG data during the individual periods in 2016. A very weak correlation was observed between wind speed and LFG data in summer, while a strong correlation was observed in winter (05.12.2016-08.12.2016). A direct correlation was observed between wind speed and LFG CH_4 concentration, while an inverse correlation was observed between wind speed and LFG flow during winter. Weak correlation between wind speed and CH_4 flow in winter 2016 ($r = 0.26$) was due to the fairly constant CH_4 flow in 2016, resulting from the automatic regulation of the gas engine when on full load, as previously discussed.

Table 7. Correlation coefficient between wind speed and barometric pressure and LFG data during the individual periods of 2016.

Period	Parameter	Barometric pressure (mbar)	LFG CH_4 concentration (%)	LFG flow (Nm^3/h)	CH_4 flow (Nm^3/h)
15.08.2016-25.08.2016	Wind speed (m/s)	0.09 (+)	-0.10 (+)	0.09 (+)	0.05 (+)
05.09.2016-11.09.2016	Wind speed (m/s)	-0.04 (+)	-0.13 (+)	0.15 (*)	0.30 (***)
05.12.2016-08.12.2016	Wind speed (m/s)	-0.91 (***)	0.75 (***)	-0.77 (***)	0.26 (**)

***: $p < 0.001$, very high significance
**: $0.01 \leq p < 0.05$, significance
*: $0.05 \leq p < 0.10$, weak significance
+: $p \geq 0.10$, no significance

Figure 14 shows wind speed against LFG CH_4 concentration, LFG flow and CH_4 flow during winter 2016 (05.12.2016-08.12.2016). Visually, it can be noted that higher wind speeds resulted in higher LFG CH_4 concentrations and lower LFG flows in winter (Figure 14), but not in summer (Figure 13). Poulsen (2005) has shown a positive influence of wind turbulence on gas emissions from landfills, especially in winter, when the soil is moister and winds are stronger. The average wind speed during winter (3.5 m/s) in this study was significantly higher than the average wind speed during summer (1.3-2.1 m/s). No information about the moisture content of the cover soils was available in this study.

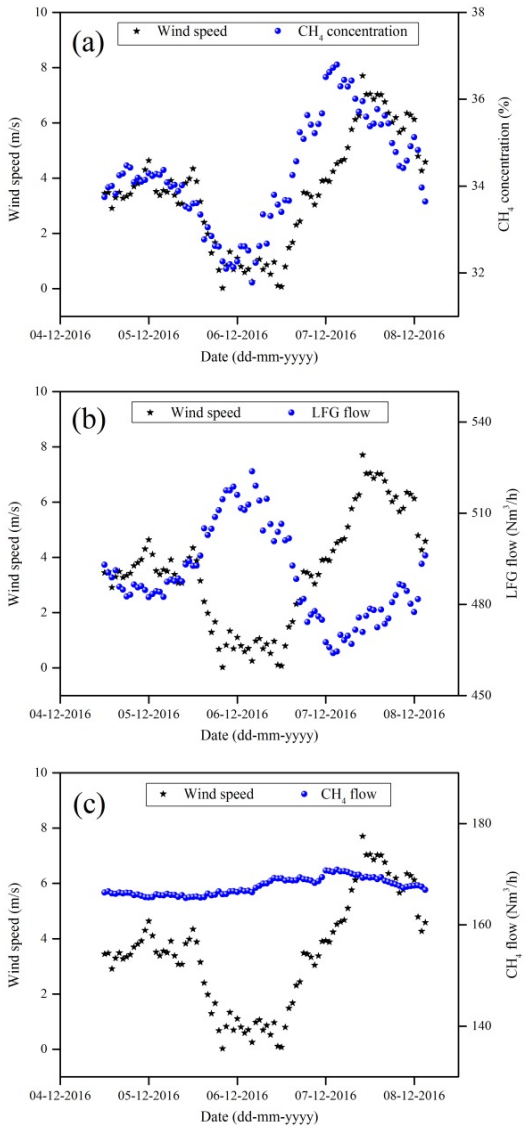

Figure 14. Wind speed (m/s) against LFG CH_4 concentration (a), LFG flow (b) and CH_4 flow (c) in the period 05.12.2016-08.12.2016.

A strong and inverse correlation was observed between wind speed and barometric pressure in winter 2016, but not in summer 2016 (Table 7). The cause-and-effect relationship between these two parameters was not investigated in this study.

Wind-induced advection has been suggested as the main CH_4 emission mechanism under windy conditions at landfills in previous studies (Poulsen, 2005; Xin et al., 2016). Xin et al. (2016) observed a positive correlation between wind speed and CH_4 emissions at night in a Chinese landfill. Wind-induced advective gas transport is caused by the blowing of strong winds across irregular topography (such as a landfill), which results in the development of pressure fields on the surface (Massman et al., 1997). This

means that strong winds decrease the pressure on the surface of the landfill, resulting in a higher pressure difference between inside and the surface of the landfill, and subsequently a higher advective gas emission and lower recovery from landfills.

Moreover, advection is more important than diffusion in a low porosity medium (Poulsen, 2005). Generally, the moisture content of cover soils is higher in winter due to higher precipitation, and thus the cover soils have lower permeability. Winds are also usually stronger in winter. Therefore, stronger winds coinciding with moister soil covers may result in wind-induced advection.

3.3.4 Solar radiation

LFG data correlated very weakly with solar radiation (Table 6). Figure 15 shows solar radiation against LFG CH_4 concentration, LFG flow and CH_4 flow in the period 11.08.2015-06.09.2015, indicating no visual correlation between solar radiation and LFG data. Performing correlation tests on the three periods in 2016 individually also showed very weak correlation between solar radiation and LFG data ($|r| = 0\text{-}0.21$). This shows that solar radiation does not affect gas recovery at these landfills.

Temperature, which affects diffusional gas transport, is affected by solar radiation. However, at landfills with active gas recovery system diffusion is less important because the main mechanism of gas transport is advection due to the applied suction pressure of the gas recovery system (Xu et al., 2014). Moreover, CH_4 oxidation and emissions at landfills are affected by soil temperature (Börjesson and Svensson, 1997; Scheutz et al., 2009), which in turn is influenced by solar radiation. However, as previously discussed, the temperature inside landfills, which affects CH_4 production, is usually higher than soil and ambient temperature due to the entrapment of produced heat by anaerobic digestion of the waste inside the landfill. This explains the lack of correlation between solar radiation and LFG data.

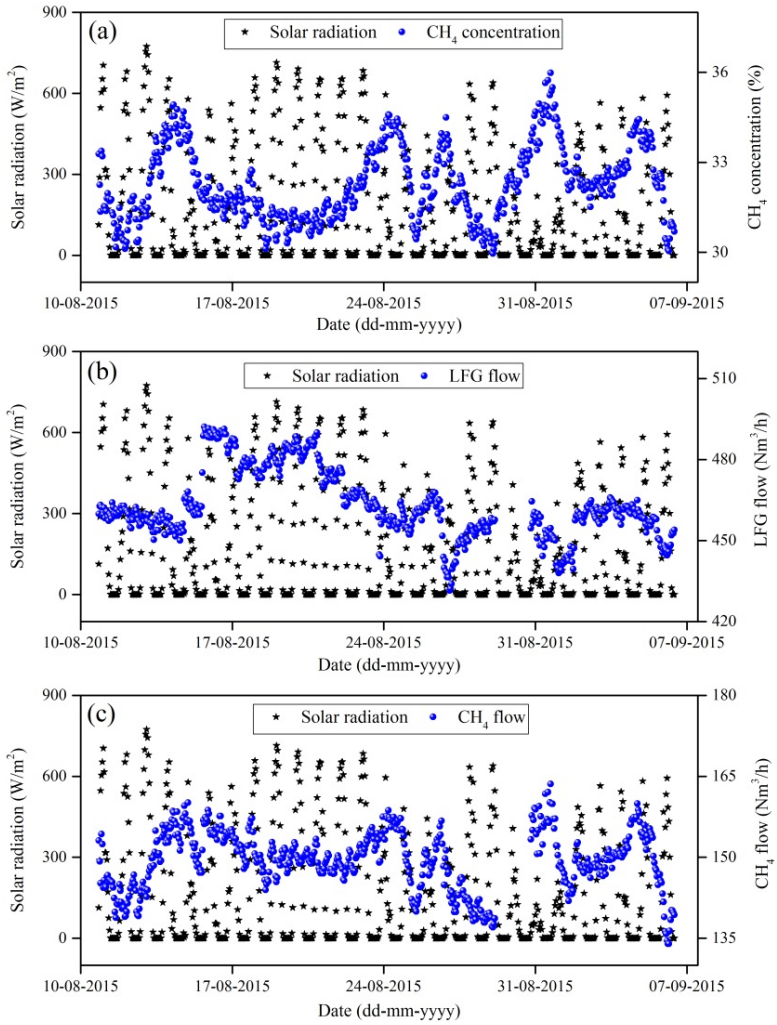

Figure 15. Solar radiation against LFG CH_4 concentration (a), LFG flow (b) and CH_4 flow (c) in the period 11.08.2015-06.09.2015.

4 Conclusions and perspectives

The objectives of this PhD project were to assess CH_4 production from shredder waste (SW) at landfills, determine gas recovery efficiency (GRE) at two adjacent Danish landfills, and investigate the impact of meteorological parameters (barometric pressure, wind speed, ambient temperature and solar radiation) on gas recovery from landfills. The objectives were met by performing laboratory experiments, field measurements, modelling and statistical analysis. The main conclusions of this project are listed below:

- High CH_4 production from SW at landfills is most probably because methanogens convert the existing CO_2 in the produced landfill gas (LFG) into CH_4, using the H_2 produced by the biocorrosion of aluminium and zinc. Moreover, the fine fraction of SW was found to have a lower BMP and k-value than the unsieved SW, meaning that landfilling of the fine fraction of SW can result in lower CH_4 production. Furthermore, temperature and moisture were found to be important factors significantly affecting the CH_4 production rate from SW.
- The GRE at the landfills, determined by field measurements, was found to be 59-76%. This showed a high potential for further improvement of the gas recovery system. The CH_4 screening results, showing the areas with high CH_4 emissions, can be utilized to develop a plan for improving gas recovery from the landfills. A good agreement was observed between the measured and modelled total CH_4 production rates (147 and 154 kg CH_4/h, respectively).
- Wind speed in winter, the absolute value of barometric pressure and changes in barometric pressure showed an influence on LFG recovery, while ambient temperature and solar radiation did not appear to be important factors influencing LFG recovery. In order to achieve higher CH_4 recovery and lower emissions, it is recommended that barometric pressure and wind speed are taken into account when regulating the gas recovery system.

5 Future research

Based on the findings of this PhD study, the following research is recommended in order to further understand the processes that affect gas production and recovery at landfills:

- The reason for high CH_4 production and the unusual gas composition from SW monofills was investigated in this work. However, the reason for high temperatures observed in SW monofills remains unknown. Thus, further research is suggested to find out the reason for these high temperatures.
- The performed CH_4 screening and emission measurements revealed the areas with high CH_4 emissions. Focusing on these revealed areas for the optimization of the gas recovery system is recommended, as well as determining the GRE after the optimization of the system and comparing it with the measured GRE in this study as the baseline, in order to assess the effectiveness of the optimization plan.
- The results of univariate data analysis showed that wind speed in winter and barometric pressure affect CH_4 recovery from landfills. However, the simultaneous impact of these two parameters on CH_4 recovery was not studied. Thus, a multivariate data analysis is further suggested in order to investigate the simultaneous impact of wind speed and barometric pressure on CH_4 recovery at landfills.
- The statistical analysis results showed that barometric pressure and wind speed highly correlate in winter. However, the cause-and-effect relationship between these two parameters was not studied. Thus, further research is recommended in order to investigate this relationship between wind speed and barometric pressure.
- In order to further understand the impact of barometric pressure and wind speed on landfill CH_4 mass balances, performing whole-site emission and oxidation measurements during periods of changes in wind speed or barometric pressure, alongside monitoring the CH_4 recovery rate, is recommended.

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7 Papers

I Aghdam, E.F., Scheutz, C., Kjeldsen, P., 2017. Assessment of methane production from shredder waste in landfills: The influence of temperature, moisture and metals. *Waste Management* 63, 226-237.

II Aghdam, E.F., Fredenslund, A.M., Chanton, J., Kjeldsen, P., Scheutz, C., 2018. Determination of gas recovery efficiency at two Danish landfills by performing downwind methane measurements and stable carbon isotopic analysis. *Waste Management* 73, 220-229.

III Aghdam, E.F., Scheutz, C., Kjeldsen, P., 2018. Impact of meteorological parameters on extracted landfill gas composition and flow. *Waste Manage*ment, in press. doi:10.1016/j.wasman.2018.01.045.

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